

# PATENT ABSTRACTS OF JAPAN

(11)Publication number : 2003-221216

(43)Date of publication of application : 05.08.2003

(51)Int.Cl.

C01B 31/02

(21)Application number : 2002-294824

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(22)Date of filing : 08.10.2002

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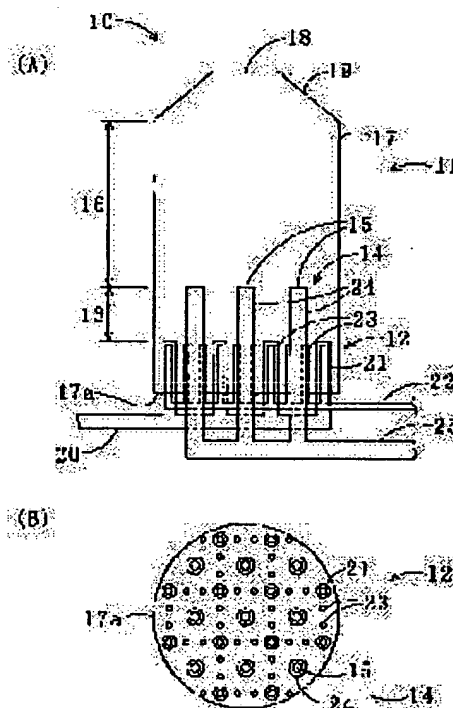
Priority number : 2001357008 Priority date : 22.11.2001 Priority country : JP

## (54) METHOD AND APPARATUS FOR MANUFACTURING FULLERENES

(57)Abstract:

**PROBLEM TO BE SOLVED:** To provide a method and an apparatus for manufacturing fullerenes economically on a large scale in a mass production.

**SOLUTION:** In an apparatus 10 for manufacturing fullerenes which apparatus has the first reaction zone 13 that forms a high temperature combustion gas flow by supplying an oxygen-containing gas and a fuel gas via the first burner 12 into a reaction furnace 11 and burning them, and the second reaction zone 16 that is situated in the downstream side of the first reaction zone 13, has discharge spouts 15 of the second burner 14 for supplying the raw material hydrocarbons into the combustion gas flow and produces fullerenes by reacting the raw material hydrocarbons supplied in a gasified state in the combustion gas flow, the apparatus is characterized in that the discharge spouts 15 of the second burner 14 are formed in the upstream side of the second reaction zone 16 in great numbers and with clearances therebetween and dispersively discharge the raw material



hydrocarbons into the combustion gas flow.

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## LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]

[Date of final disposal for application]

[Patent number]

[Date of registration]

[Number of appeal against examiner's decision of rejection]

[Date of requesting appeal against examiner's decision of rejection]

[Date of extinction of right]

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(19) 日本国特許庁 (J P)

(12) 公開特許公報 (A)

(11) 特許出願公開番号

特開2003-221216

(P2003-221216A)

(43) 公開日 平成15年8月5日(2003.8.5)

(51) Int.Cl.<sup>7</sup>

C 01 B 31/02

識別記号

1 0 1

F I

C 01 B 31/02

テームコード(参考)

1 0 1 F 4 G 1 4 6

審査請求 未請求 請求項の数13 O L (全 14 頁)

(21) 出願番号 特願2002-294824(P2002-294824)

(22) 出願日 平成14年10月8日(2002.10.8)

(31) 優先権主張番号 特願2001-357008(P2001-357008)

(32) 優先日 平成13年11月22日(2001.11.22)

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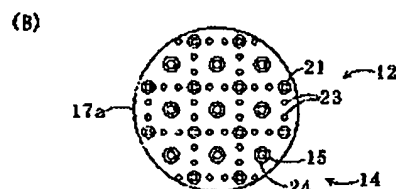
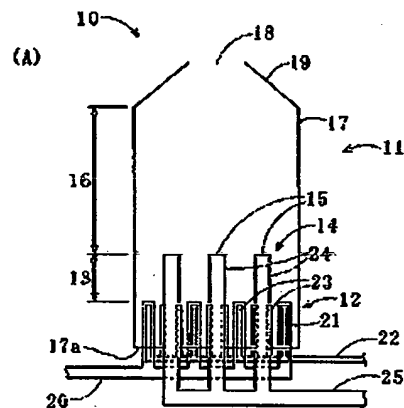
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(54) 【発明の名称】 フラレーン類の製造方法及びその装置

(57) 【要約】

【課題】 経済的かつ大量生産可能なフラレーン類の製造方法及びその装置を提供する。

【解決手段】 反応炉11内に、第1のバーナー12を介して酸素含有ガスと燃料ガスを供給し、燃焼させて高温の燃焼ガス流を形成させる第1反応帯域13と、第1反応帯域13の下流側にあつて、燃焼ガス流に原料炭化水素を供給する第2のバーナー14の吐出口15を有し、ガス化して供給された原料炭化水素を燃焼ガス流中で反応させてフラレーン類を生成させる第2反応帯域16を有するフラレーン類の製造装置10において、第2のバーナー14の吐出口15は、第2反応帯域16の上流側に隙間を有して多数形成され、原料炭化水素を燃焼ガス流中に分散放出する。



## 【特許請求の範囲】

【請求項1】 反応炉内に、酸素含有ガスと燃料とを供給して燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、この燃焼ガス流の途中に原料炭化水素を供給する原料炭化水素供給口を有し且つ該原料炭化水素を反応させてフラーレン類を生成させる第2反応帯域を有することを特徴とするフラーレン類の製造装置を使用し、前記第2反応帯域の圧力を大気圧未満とすることを特徴とするフラーレン類の製造方法。

【請求項2】 請求項1に記載のフラーレン類の製造方法において、前記第2反応帯域が前記第1反応帯域の下流側にあることを特徴とするフラーレン類の製造方法。

【請求項3】 請求項1及び2のいずれか1項に記載のフラーレン類の製造方法において、前記第2反応帯域の温度が1000℃以上であることを特徴とするフラーレン類の製造方法。

【請求項4】 反応炉内に、第1のバーナーを介して酸素含有ガスと燃料ガスを供給し、これらを燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、該第1反応帯域の下流側において、前記燃焼ガス流に原料炭化水素を供給する第2のバーナーの吐出口を有し、ガス化して供給された前記原料炭化水素を前記燃焼ガス流中で反応させてフラーレン類を生成させる第2反応帯域を有することを特徴とするフラーレン類の製造装置。

【請求項5】 請求項4記載のフラーレン類の製造装置において、前記第2のバーナーの吐出口は、前記第2反応帯域の上流側に隙間を有して多数形成され、前記原料炭化水素を前記燃焼ガス流中に分散放出することを特徴とするフラーレン類の製造装置。

【請求項6】 請求項5記載のフラーレン類の製造装置において、前記第2のバーナーは、前記第1反応帯域を貫通して配置される多数の小径吐出管からなっていることを特徴とするフラーレン類の製造装置。

【請求項7】 請求項4～6のいずれか1項に記載のフラーレン類の製造装置において、前記第1のバーナーは、前記酸素含有ガスと前記燃料ガスをそれぞれ独立に放出する複数の酸素含有ガスノズル及び燃料ガスノズルが混在配置されていることを特徴とするフラーレン類の製造装置。

【請求項8】 請求項4～6のいずれか1項に記載のフラーレン類の製造装置において、前記第1のバーナーのヘッドは多孔質部材からなっており、表面から前記酸素含有ガスと前記燃料ガスが混合された状態で噴出されることを特徴とするフラーレン類の製造装置。

【請求項9】 請求項8記載のフラーレン類の製造装置において、前記酸素含有ガスと前記燃料ガスの混合は前記第1のバーナー内で行われ、前記第1のバーナーには前記酸素含有ガスと前記燃料ガスが独立に別配管で供給されていることを特徴とするフラーレン類の製造装置。

【請求項10】 請求項8記載のフラーレン類の製造装置

において、前記酸素含有ガスと前記燃料ガスとは予混合されて前記ヘッドの下部に設けられた蓄圧室に供給されていることを特徴とするフラーレン類の製造装置。

【請求項11】 請求項4～6のいずれか1項に記載のフラーレン類の製造装置において、前記第1のバーナーは、多数の小径の噴出ノズルが隙間において形成されたヘッダー管を有し、該ヘッダー管には予混合された前記酸素含有ガスと前記燃料ガスが供給されていることを特徴とするフラーレン類の製造装置。

【請求項12】 請求項4～6のいずれか1項に記載のフラーレン類の製造装置において、前記第1のバーナーは、前記酸素含有ガスを噴出する多数の小径の噴出ノズルが隙間において形成された第1のヘッダー管と、前記第1のヘッダー管とは隙間を有し配置され前記燃料ガスを噴出する多数の小径の噴出ノズルが隙間において形成された第2のヘッダー管を有し、前記第1のヘッダー管及び前記第2のヘッダー管には前記酸素含有ガス及び前記燃料ガスがそれぞれ独立に別配管で供給されていることを特徴とするフラーレン類の製造装置。

【請求項13】 請求項4～12のいずれか1項に記載のフラーレン類の製造装置において、前記第2のバーナーから供給される原料炭化水素に酸素含有ガスを混合することを特徴とするフラーレン類の製造装置。

## 【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、フラーレン類の製造方法及びその装置に関する。

【0002】

【従来の技術】フラーレン類（以下、単にフラーレンとすることもある）は、ダイヤモンド、黒鉛に次ぐ第三の炭素同素体の総称であり、 $C_{60}$ 、 $C_{70}$ 等に代表されるように5員環と6員環のネットワークで閉じた中空殻状の炭素分子である。フラーレンの存在が最終的に確認されたのは比較的最近の1990年のことであり、比較的新しい炭素材料であるが、その特殊な分子構造ゆえに特異的な物理的性質を示すことが認められ、例えば、以下のような広範囲な分野に渡り、革新的な用途開発が急速に展開されつつある。

（1）超硬材料への応用：フラーレンを前駆体とすることで微細結晶粒子をもつ人工ダイヤモンドの精製が可能のため、付加価値のある耐磨耗材料への利用が期待されている。

（2）医薬品への応用： $C_{60}$ 、誘導体、光デバイスを用いることで抗癌剤、エイズ・骨粗鬆症・アルツハイマー治療薬、造影剤、ステント材料等の用途としての研究が進められている。

（3）超伝導材料への応用：フラーレン薄膜に金属カリウムをドーブすると18Kという高い転移温度を持つ超伝導材料をつくり出すことができることが発見され、多方面から注目を集めている。

(4) 半導体製造への応用：レジストにC<sub>60</sub>を混ぜること  
でレジスト構造がより一層強化されることを利用し、  
次世代半導体製造への応用が期待されている。

【0003】各種炭素数のフラーレンの中でもC<sub>60</sub>及び  
C<sub>70</sub>は比較的合成が容易であり、それゆえ今後の需要も  
爆発的に高まることが予想されている。現在知られてい  
るフラーレンの製造方法としては、以下に示す方法が挙  
げられる。

#### (1) レーザ蒸着法

希ガス中に置かれた炭素ターゲットに高エネルギー密度  
のバルスレーザーを照射し、炭素原子の蒸発により合成  
する方法である。希ガスが流れる石英管を電気炉の中に  
置き、グラファイト試料をその石英管の中に置く。ガス  
の流れの上流側からグラファイト試料にレーザーを照射  
し、蒸発させると電気炉出口付近の冷えた石英管の内壁  
にC<sub>60</sub>やC<sub>70</sub>などのフラーレンを含む炭が付着する。レ  
ーザーのショット当たりの蒸発量がわずかで、大量  
製造には不向きである。

#### (2) 抵抗加熱法

ヘリウムガスで満たされた減圧下の容器の中でグラファ  
イト棒を通過加熱し昇華させる方法である。回路での電  
気抵抗ロスが大きいため大量製造に不向きである。

#### 【0004】(3) アーク放電法

数十kPa中のヘリウムガス中で2本のグラファイト電  
極を軽く接触させたり、あるいは1～2mm程度離れた  
状態でアーク放電を起こし、陽極の炭素を昇華させる方  
法である。現在工場規模でのフラーレンの大量製造に用  
いられている。

#### (4) 高周波誘導加熱法

抵抗加熱やアーク放電を併用代りに、高周波誘導により  
原料グラファイトに渦電流を流し、原料グラファイトを  
加熱して蒸発させる方法である。

#### (5) 燃焼法

ヘリウム等の不活性ガスと酸素との混合ガス中でベンゼ  
ン等の炭化水素原料を不完全燃焼させる方法である。ベン  
ゼン燃料の燃%が煤となり、その10%程度がフラー  
レンとなる点で製造効率はよくないが、副製する煤(フ  
ラーレン等)を液体燃料等に使用可能な点、製造装置が  
単純である点で、アーク放電法に対抗する大量生産法と  
して注目されている。

#### (6) ナフタレン熱分解法

ナフタレンを約1000℃で熱分解させる方法である。  
【0005】このように現在までにさまざまなフラーレン  
の合成法が提案されているが、いずれの方法によっても  
これまでにフラーレンを安価に大量に製造する方法は  
確立されていない。これらの方法のうち、最も安価で、  
効率的な製造方法の一つと考えられるのは燃焼法であ  
り、例えば、特許文献1には、炭素含有物を火炎中で燃  
焼させ凝縮物を収集することによるフラーレンの製造方  
法が記載されている。この方法は、炭素含有物を火炎中

で燃焼させることによりフラーレンを製造する方法であ  
り、実質的に燃焼のための燃料とフラーレンの原料は同  
一の炭素含有物である。フラーレンは煤状物質中に含ま  
れて生成されるが、この煤状物質の一部はいわゆるカー  
ボンブラックである。

【0006】カーボンブラックの製造方法としては、フ  
ァーネス法、チャンネル法、サーマル法、アセチレン法  
などが知られており、工業的に一般的な製造方法として  
はファーネス法が挙げられる。この方法は、例えば円筒  
状のカーボンブラック製造装置(反応炉)を使用し、当  
該反応炉の第1反応帯域に炉軸に対して水平方向又は垂  
直方向に空気などの酸素含有ガスと燃料を供給し且つ燃  
焼させ、得られた燃焼ガス流を炉軸方向の下流に設置さ  
れ縮小した断面積を持つ第2反応帯域に移動させ、当該  
ガス流中に原料炭化水素(原料油)を供給し反応させて  
カーボンブラックを生成させ、更に、その下流にある第  
3反応帯域でガス流に冷却水の噴霧などでガスを急冷し  
て反応を停止させる方法である。

#### 【0007】

【特許文献1】特表平6-507879号公報

#### 【0008】

【発明が解決しようとする課題】しかしながら、上記の  
通常のカーボンブラックの製造方法では、フラーレンは  
ほとんど生成しない。フラーレンの製造においては、得  
られる煤状物質中に含まれるフラーレンの割合をいかに  
高めるかが大きな課題となっている。一般的に、フラー  
レンの製造は、減圧下で行われ、反応領域中に希釈剤を  
導入する場合もある。これらの減圧度、希釈剤濃度は上  
記フラーレンの収率に影響を及ぼすことが知られてい  
る。

【0009】上記、特許文献1にはフラーレンの収率を  
向上させるために、火炎温度を上昇させること、またそ  
の手段として外部エネルギー源から火炎にさらにエネル  
ギーを供給する方法が述べられている。好ましいエネル  
ギー源としては、火炎を直接加熱する電気抵抗加熱、マ  
イクロウェーブ加熱、放電加熱、及び高温ガスとの熱交  
換で火炎を加熱する向流加熱が挙げられている。

【0010】上記、特許文献1では、燃焼反応のための  
酸化剤として純酸素が、また希釈剤としてはアルゴンが  
用いられている。これは、フラーレンの収率を上げる効  
果があると考えられる。しかしながら、純酸素は専用の  
ボンベもしくは供給設備等を要し、特に、工業規模でフ  
ラーレンを製造しようとする場合には、燃焼のために必  
要とする酸素の量も大量になり、特別な酸素供給設備が  
必要で、結果としてフラーレンの製造コストも高価とな  
る。

【0011】そこで、燃焼法において製造コストを低減  
するために、燃焼の酸化剤として空気を用いることは容  
易に類推できるが、純酸素に比べて酸素濃度が低いた  
めに火炎が安定しないことや窒素の割合が多いため燃焼温

度が低くなること、特に減圧下での操作時に体積が増えノズルを通過する速度が速くなる等の理由により実用化には至っていない。フラーレンは次世代を担う新材料、新素材として多方面から注目されており、フラーレンを大量に且つ安価に、そして容易に製造する技術の開発が望まれている。

【0012】本発明は前述したような事情に鑑みてなされたものであり、フラーレンを大量に且つ安価に、そして容易に製造するフラーレン類の製造方法及びその装置を提供することを目的とする。

【0013】

【課題を解決するための手段】本発明者らは、フラーレンを大量に且つ安価に製造できる最適な燃焼方法及び製造装置を種々検討した結果、反応炉内に、酸素含有ガスと燃料とを供給し且つ燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、燃焼ガス流に原料炭化水素を供給する原料炭化水素供給口を有し且つ原料炭化水素を反応させてフラーレンを生成させる第2反応帯域を有するフラーレン類の製造装置を用いて、第2反応帯域の圧力を大気圧未満に保つことでフラーレンを大量に安定的に生成できるとの知見を得た。

【0014】即ち、前記目的に沿う第1の発明に係るフラーレン類の製造方法は、反応炉内に、酸素含有ガスと燃料とを供給して燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、この燃焼ガス流の途中に原料炭化水素を供給する原料炭化水素供給口を有し且つ原料炭化水素を反応させてフラーレン類を生成させる第2反応帯域を有することを特徴とするフラーレン類の製造装置を使用し、前記第2反応帯域の圧力を大気圧未満とする。第1反応帯域に燃料と酸素含有ガスを供給して燃焼させるので、例えば完全燃焼を容易に達成することができる。高温の燃焼ガス流を形成することができる。そして、得られた高温のガス流中に原料炭化水素を供給することにより、原料炭化水素を容易に熱分解させることができる。また、第2反応帯域内の圧力を大気圧未満にして、原料炭化水素と燃焼ガスの混合状態を希薄にすることにより、原料炭化水素の熱分解が均一に進行して、フラーレン類の生成効率を向上させることができる。

【0015】第1の発明に係るフラーレン類の製造方法において、前記第2反応帯域が前記第1反応帯域の下流側にあることが好ましい。第2反応帯域を第1反応帯域の下流側に設けることにより、第1反応帯域で形成された高温の燃焼ガスを直ちに第2反応帯域に導入することができる。その結果、第2反応帯域の温度を高温にすることができる。第1の発明に係るフラーレン類の製造方法において、前記第2反応帯域の温度が1000℃以上であることが好ましい。第2反応帯域の温度を1000℃以上にするにより、供給された原料炭化水素を短時間に確実に熱分解させることができる。

【0016】前記目的に沿う第2の発明に係るフラーレン類の製造装置は、反応炉内に、第1のバーナーを介して酸素含有ガスと燃料ガスを供給し、これらを燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、該第1反応帯域の下流側にあって、前記燃焼ガス流に原料炭化水素を供給する第2のバーナーの吐出口を有し、ガス化して供給された前記原料炭化水素を前記燃焼ガス流中で反応させてフラーレン類を生成させる第2反応帯域を有する。燃料の燃焼は第1反応帯域で行なうので、燃焼状態の制御が容易となり、高温の燃焼ガスを容易に形成することができる。得られた高温の燃焼ガス流を第2反応帯域に導入し、この高温のガス流中に原料炭化水素を供給して熱分解させるので、高温の燃焼ガス流の温度、流速、流量等のガス流条件と原料炭化水素の供給条件を調整することにより、原料炭化水素の熱分解の制御が容易となる。

【0017】第2の発明に係るフラーレン類の製造装置において、前記第2のバーナーの吐出口は、前記第2反応帯域の上流側に隙間を有して多数形成され、前記原料炭化水素を前記燃焼ガス流中に分散放出することが好ましい。原料炭化水素を供給する第2のバーナーの吐出口を第2反応帯域の上流側に形成することにより、第1反応帯域から流入する高温の燃焼ガス流中に直接原料炭化水素を供給することができ、原料炭化水素を容易に熱分解させることができる。また、原料炭化水素を多数の吐出口から燃焼ガス中に分散放出するので、燃焼ガス中で原料炭化水素を短時間に均一に熱分解することができる。第2の発明に係るフラーレン類の製造装置において、前記第2のバーナーは、前記第1反応帯域を貫通して配置される多数の小径吐出管からなっていることが好ましい。多数の小径吐出管で原料炭化水素が供給されるため、第2反応帯域の高温の燃焼ガス流中に原料炭化水素を一様に分散放出することができる。また、小径吐出管は第1反応帯域を貫通して配置されているので、原料炭化水素は小径吐出管内を通過しながら高温の燃焼ガスにより徐々に加熱されて、第2反応帯域の高温の燃焼ガス流中で熱分解を促進することができる。

【0018】第2の発明に係るフラーレン類の製造装置において、前記第1のバーナーは、前記酸素含有ガスと前記燃料ガスをそれぞれ独立に放出する複数の酸素含有ガスノズル及び燃料ガスノズルが混在配置されているもよい。このような構成とすることにより、供給された酸素含有ガスと燃料ガスは並流混合して一様な混合状態となって第1反応帯域に存在させることができる。また、第2の発明に係るフラーレン類の製造装置において、前記第1のバーナーのヘッドは多孔質部材からなっており、表面から前記酸素含有ガスと前記燃料ガスが混合された状態で噴出される構成とすることができる。このような構成とすることにより、酸素含有ガスと燃料ガスを予混合された状態で第1反応帯域に供給することができ

度が低くなること、特に減圧下での操作時に体積が増えノズルを通過する速度が速くなる等の理由により実用化には至っていない。フラーレンは次世代を担う新材料、新素材として多方面から注目されており、フラーレンを大量に且つ安価に、そして容易に製造する技術の開発が望まれている。

【0012】本発明は前述したような事情に鑑みてなされたものであり、フラーレンを大量に且つ安価に、そして容易に製造するフラーレン類の製造方法及びその装置を提供することを目的とする。

【0013】

【課題を解決するための手段】本発明者は、フラーレンを大量に且つ安価に製造できる最適な燃焼方法及び製造装置を種々検討した結果、反応炉内に、酸素含有ガスと燃料とを供給し且つ燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、燃焼ガス流に原料炭化水素を供給する原料炭化水素供給口を有し且つ原料炭化水素を反応させてフラーレンを生成させる第2反応帯域を有するフラーレン類の製造装置を用いて、第2反応帯域の圧力を大気圧未満に保つことでフラーレンを大量に安定的に生成できるとの知見を得た。

【0014】即ち、前記目的に沿う第1の発明に係るフラーレン類の製造方法は、反応炉内に、酸素含有ガスと燃料とを供給して燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、この燃焼ガス流の途中に原料炭化水素を供給する原料炭化水素供給口を有し且つ該原料炭化水素を反応させてフラーレン類を生成させる第2反応帯域を有することを特徴とするフラーレン類の製造装置を使用し、前記第2反応帯域の圧力を大気圧未満とする。第1反応帯域に燃料と酸素含有ガスを供給して燃焼させるので、例えば完全燃焼を容易に達成することができる。高温の燃焼ガス流を形成することができる。そして、得られた高温のガス流中に原料炭化水素を供給することにより、原料炭化水素を容易に熱分解させることができ、フラーレン類の生成効率を向上させることができる。また、第2反応帯域内の圧力を大気圧未満にして、原料炭化水素と燃焼ガスの混合状態を希薄にすることにより、原料炭化水素の熱分解が均一に進行して、フラーレン類の生成効率を向上させることができる。

【0015】第1の発明に係るフラーレン類の製造方法において、前記第2反応帯域が前記第1反応帯域の下流側にあることが好ましい。第2反応帯域を第1反応帯域の下流側に設けることにより、第1反応帯域で形成された高温の燃焼ガスを直ちに第2反応帯域に導入することができる。その結果、第2反応帯域の温度を高温にすることができる。第1の発明に係るフラーレン類の製造方法において、前記第2反応帯域の温度が1000℃以上であることが好ましい。第2反応帯域の温度を1000℃以上にするにより、供給された原料炭化水素を短時間に確実に熱分解させることができる。

【0016】前記目的に沿う第2の発明に係るフラーレン類の製造装置は、反応炉内に、第1のバーナーを介して酸素含有ガスと燃料ガスを供給し、これらを燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、該第1反応帯域の下流側において、前記燃焼ガス流に原料炭化水素を供給する第2のバーナーの吐出口を有し、ガス化して供給された前記原料炭化水素を前記燃焼ガス流中で反応させてフラーレン類を生成させる第2反応帯域を有する。燃料の燃焼は第1反応帯域で行なうので、燃焼状態の制御が容易となり、高温の燃焼ガスを容易に形成することができる。得られた高温の燃焼ガス流を第2反応帯域に導入し、この高温のガス流中に原料炭化水素を供給して熱分解させるので、高温の燃焼ガス流の温度、流速、流量等のガス流条件と原料炭化水素の供給条件を調整することにより、原料炭化水素の熱分解の制御が容易となる。

【0017】第2の発明に係るフラーレン類の製造装置において、前記第2のバーナーの吐出口は、前記第2反応帯域の上流側に隙間を有して多数形成され、前記原料炭化水素を前記燃焼ガス流中に分散放出することが好ましい。原料炭化水素を供給する第2のバーナーの吐出口を第2反応帯域の上流側に形成することにより、第1反応帯域から流入する高温の燃焼ガス流中に直接原料炭化水素を供給することができ、原料炭化水素を容易に熱分解させることができる。また、原料炭化水素を多数の吐出口から燃焼ガス中に分散放出するので、燃焼ガス中で原料炭化水素を短時間に均一に熱分解することができる。第2の発明に係るフラーレン類の製造装置において、前記第2のバーナーは、前記第1反応帯域を貫通して配置される多数の小径吐出管からなっていることが好ましい。多数の小径吐出管で原料炭化水素が供給されるため、第2反応帯域の高温の燃焼ガス流中に原料炭化水素を均一に分散放出することができる。また、小径吐出管は第1反応帯域を貫通して配置されているので、原料炭化水素は小径吐出管内を通過しながら高温の燃焼ガスにより徐々に加熱されて、第2反応帯域の高温の燃焼ガス流中で熱分解を促進することができる。

【0018】第2の発明に係るフラーレン類の製造装置において、前記第1のバーナーは、前記酸素含有ガスと前記燃料ガスをそれぞれ独立に放出する複数の酸素含有ガスノズル及び燃料ガスノズルが混在配置されていてもよい。このような構成とすることにより、供給された酸素含有ガスと燃料ガスは並流混合して一様な混合状態となって第1反応帯域に存在させることができる。また、第2の発明に係るフラーレン類の製造装置において、前記第1のバーナーのヘッドは多孔質部材からなっており、表面から前記酸素含有ガスと前記燃料ガスが混合された状態で噴出される構成とすることができる。このような構成とすることにより、酸素含有ガスと燃料ガスを予混合された状態で第1反応帯域に供給することができ

る。

【0019】第2の発明に係るフラーレン類の製造装置において、前記酸素含有ガスと前記燃料ガスの混合は前記第1のバーナー内で行われ、前記第1のバーナーには前記酸素含有ガスと前記燃料ガスが独立に別配管で供給される構成とすることができる。酸素含有ガスと燃料ガスの混合が第1のバーナー内で行われるので、酸素含有ガスと燃料ガスとの予混合手段を別個に設ける必要がなく、フラーレン類の製造装置の構成が簡単となる。第2の発明に係るフラーレン類の製造装置において、前記酸素含有ガスと前記燃料ガスとは予混合されて前記ヘッド

の下部に設けられた蒼圧室に供給される構成とすることができる。酸素含有ガスと燃料ガスが予混合されてヘッドの下部の蒼圧室に供給されるので、第1のバーナーの構造を簡単とすることができる。

【0020】第2の発明に係るフラーレン類の製造装置において、前記第1のバーナーは、多数の小径の噴出ノズルが隙間において形成されたヘッダー管を有し、該ヘッダー管には予混合された前記酸素含有ガスと前記燃料ガスが供給される構成とすることができる。このような構成とすることにより、酸素含有ガスと燃料ガスを予混合された状態で第1反応帯域に分散放出することができる。第2の発明に係るフラーレン類の製造装置において、前記第1のバーナーは、前記酸素含有ガスを噴出する多数の小径の噴出ノズルが隙間において形成された第1のヘッダー管と、前記第1のヘッダー管とは隙間を有し配置され前記燃料ガスを噴出する多数の小径の噴出ノズルが隙間において形成された第2のヘッダー管を有し、前記第1のヘッダー管及び前記第2のヘッダー管には前記酸素含有ガス及び前記燃料ガスがそれぞれ独立に別配管で供給される構成とすることができる。このような構成とすることにより、分散放出された酸素含有ガスと燃料ガスは並散混合して一様な混合状態となって第1反応帯域に存在させることができる。

【0021】第2の発明に係るフラーレン類の製造装置において、前記第2のバーナーから供給される原料炭化水素に酸素含有ガスを混合することができる。原料炭化水素の熱分解は吸熱反応のため、原料炭化水素の熱分解により燃焼ガスの温度は低下する。このため、原料炭化水素に酸素含有ガスを混合することにより第2反応帯域で原料炭化水素の一部を燃焼させて熱エネルギーを発生させ、原料炭化水素が熱分解する際に消費した熱エネルギーを補填して燃焼ガスの温度が低下するのを防止できる。

【0022】

【発明の実施の形態】 続いて、添付した図面を参照しつつ、本発明を具体化した実施の形態につき説明し、本発明の理解に供する。ここに、図1(A)、(B)はそれぞれ本発明の第1の実施の形態に係るフラーレン類の製造方法を適用したフラーレン製造装置の説明図、平断面

図、図2(A)、(B)はそれぞれ本発明の第2の実施の形態に係るフラーレン類の製造装置の説明図、平断面図、図3(A)、(B)はそれぞれ本発明の第3の実施の形態に係るフラーレン類の製造装置の説明図、平断面図、図4は本発明の第4の実施の形態に係るフラーレン類の製造装置の部分説明図、図5(A)、(B)はそれぞれ本発明の第5の実施の形態に係るフラーレン類の製造装置の説明図、平断面図、図6(A)、(B)はそれぞれ本発明の第6の実施の形態に係るフラーレン類の製造装置の説明図、平断面図である。

【0023】本発明の第1の実施の形態に係るフラーレン類の製造方法について、図1を用いて説明する。第1の実施の形態に係るフラーレン類の製造方法は、反応炉3a内に第1反応帯域1及び第2反応帯域2を設けて構成されるフラーレン類の製造装置3に、原料炭化水素を導入し、燃焼することによりフラーレンを製造する方法に関するものである。

【0024】フラーレン類の製造装置3は、燃焼ガス流を形成させる第1反応帯域1、そこで形成された燃焼ガス流に原料炭化水素を供給し、反応させてフラーレンを生成させる第2反応帯域2を有する。第2反応帯域2は、第1反応帯域1とはほぼ同じ領域（外側もしくは内側）であってもよく、また第1反応帯域1で形成された燃焼ガス流方向（以下、「軸方向」ということがある。）の下流側にあってもよい。

【0025】図1は、第2反応帯域2が第1反応帯域1の下流にある場合を示している。

【第1反応帯域について】 第1反応帯域1では、一般に燃料供給口及び酸素含有ガス供給口からそれぞれ燃料及び酸素含有ガスを供給し、燃焼させることで高温の燃焼ガス流を第2反応帯域2、すなわち反応炉3aの下流に向かって発生させる。

【0026】燃料及び酸素含有ガスの供給は、反応炉3a内に入る前に混合する、いわゆる予混合方式であっても、それぞれ独立したノズルから反応炉3aに供給する、いわゆる並散混合方式であってもよい。並散混合方式の場合は、図1において、例えば、中央の燃料供給口7から燃料を供給し、その周囲の酸素含有ガス供給口5、6から酸素含有ガスを供給する。また、予混合方式と並散混合方式を組み合わせてもよい。例えば、図1において、酸素含有ガス供給口5からは、燃料と酸素含有ガスをあらかじめ混合させたものを供給し、酸素含有ガス供給口6からは酸素含有ガスを、燃料供給口7からは燃料をそれぞれ独立に供給してもよい。

【0027】この第1反応帯域1は、高温の燃焼ガスを発生させることが目的であり、その燃焼方法は予混合燃焼、並散燃焼、層流燃焼、乱流燃焼、高温空気燃焼等、公知のいかなる燃焼方法であってもよい。また、第2反応帯域2でフラーレンの生成が可能となる温度が得られれば、第1反応帯域1での燃焼は完全燃焼であっても、



不完全燃焼であってもよいが、燃料使用量に対する発熱量が大きい完全燃焼であることが好ましい。第1反応帯域1がいわゆる燃料過剰の不完全燃焼である場合は、第1反応帯域1でもフラレーンを含む炭状物質が生成することがある。

【0028】しかし、好ましくはこの第1反応帯域1における燃焼は、燃焼に必要な酸素が、理論酸素量以上である。希薄混合気での燃焼の方がよい。酸素含有ガスとしては空気、酸素ガス又はこれらにアルゴンガス、窒素ガス等の不燃性ガスを任意の割合で混合したガスを使用することができる。特に高温燃焼における $\text{NO}_x$ の発生を抑えるためには、純酸素を使用してもよい。フラレーンの収率を上げるためには、燃焼過程において希ガス等を用いて希釈することが好ましい。希ガスは、供給用の専用ノズルから供給してもよいし、燃料、原料炭化水素、酸素含有ガス中にあらかじめ混合させておいてもよい。

【0029】燃料としては、水素、一酸化炭素、天然ガス、石油ガス等の燃料ガス、直油、ベンゼン、トルエンなどの石油系液体燃料、クレオソート油等の石炭系液体燃料を使用することができる。中でも、本実施の形態で使用する燃料としては燃料ガスが好ましい。また、フラレーン製造時の第1反応帯域1における平均温度は、得ようとする目的のフラレーンによって適宜調整すればよいが、好ましくは1300℃以上、更に好ましくは1600℃以上とされる。これは、燃焼ガスの温度が高温である程、フラレーン類の生産性が上がるからである。上限はあまり高すぎてもフラレーン類の生産性が落ちる場合がある。また、反応炉の材質による耐熱性の問題を考慮の上決定すればよい。

【0030】燃料供給口7、酸素含有ガス供給口5、6の配置は、反応炉3aに開口していれば任意である。図1においては、燃料供給口7、酸素含有ガス供給口5、6は、反応炉3aの同一側に開口している。反応炉3a内に開口している各供給口5、6、7の形状は任意であり、略円形、楕円形、三角・四角状などの多角形状やひょうたん型などの不定形であってもよい。

【0031】反応炉3a内圧力は大気圧未満であることが好ましく、より好ましい範囲は10～300torrである。

【第2反応帯域について】第2反応帯域2では第1反応帯域1で形成された燃焼ガス流に原料炭化水素を原料炭化水素供給口4から供給し、この原料炭化水素を一部部分燃焼させることによってフラレーン類を生成させる。部分燃焼させるために、酸素が残存するように第1反応帯域1における燃焼を酸素過剰としてもよい。また、第2反応帯域2にノズルを配置し、酸素含有ガス供給ノズルから酸素含有ガスを供給してもよい。

【0032】この際、燃焼ガス中に供給される上記原料炭化水素や酸素含有ガスは、極力均一に反応炉3a内に

供給されることが好ましい。このため、第2反応帯域2に設置する原料炭化水素供給口4、及び酸素含有ガス供給ノズルの本数は多いほどよく、また反応炉3a内に均等に配置されることが望ましい。

【0033】第2反応帯域の長さは、反応炉3aの大きさ、製造するフラレーンの種類などによって適宜選択すればよい。第2反応帯域の位置及び形状は、任意であり、第1反応帯域の内側であっても、外側であってもよく、図1に示すように、第1反応帯域1の下流側にあってもよい。第2反応帯域の形状も任意であるが、第2反応帯域の断面形状は変化しないほうが好ましい。その理由は、フラレーン類が生成する過程で第2反応帯域の断面形状が変化することによる流れの乱れの影響を受けると、生成するフラレーン類に好ましくない影響を与えるからである。

【0034】第2反応帯域2の平均温度は、製造するフラレーンによって適宜選択すればよいが、原料炭化水素が均一に気化、反応するために充分高温雰囲気であることが好ましい。具体的には1000℃以上であることが好ましく、中でも1000～1900℃、特に1700～1900℃であることが好ましい。また、第2反応帯域2においては、燃焼ガス中の酸素濃度をできるだけ抑制することが好ましい。燃焼ガス中に酸素が多量に存在すると、フラレーン類の生成反応帯域すなわち第2反応帯域2での原料炭化水素の一部燃焼が活発に起こり、そのため、第2反応帯域2での温度の不均一が生じることがあるからである。燃焼ガス中の酸素濃度は、好ましくは3vol%以下、更に好ましくは0.05～1vol%である。

【0035】本実施の形態においては、原料炭化水素を供給する位置は任意であり、反応炉の形状に合わせて原料炭化水素供給口を設けることができる。例えば、反応炉3aの径が最大となっている部分に原料炭化水素供給口を設けてもよく、また、径が縮小している縮小部に原料炭化水素供給口を設けてもよい。更に、図1に示すように、反応炉3aの径が最大となっている部分と径が縮小している縮小部にそれぞれ原料炭化水素供給口4を設けてもよい。原料炭化水素供給口4の位置によって、原料炭化水素が導入される位置でのガスの流速、乱流の強さなどを制御できる。

【0036】原料炭化水素としては、従来公知の任意のものを使用することができ、例えば、ベンゼン、トルエン、キシレン、ナフタレン、アントラセン等の芳香族系炭化水素、クレオソート油、カルボン酸油などの石炭系炭化水素、エチレンヘビーエンドオイル、FCCオイル（流動接触分解残油）等の石油系重質油、アセチレン系不飽和炭化水素、エチレン系炭化水素、ペンタンやヘキサン等の脂肪族飽和炭化水素などが挙げられ、これらを単独又は任意の割合で混合して使用してもよい。中でも縮合した芳香族系炭化水素を用いることが好ましく、

特にベンゼンやトルエン等の芳香族系炭化水素が好ましい。原料の純度は高い方が好ましく、中でも芳香族系炭化水素を用いる際には純度が100%に近いほどよい。

【0037】反応炉における原料炭化水素供給口の位置は、燃焼ガスの流れ方向の断面円周上に複数設けてもよく、更には、このような同一円周上に原料炭化水素供給口を複数有する場所を、燃焼ガスの流れ方向に多段に設けてもよい。フラレーンの生成反応時間を均一にし、物性が均一なフラレーンを得るためには、同一円周上になるべく多くの原料炭化水素供給口を設置するのが好ましい。

【0038】また、原料炭化水素供給口4に使用するノズルの型式は適宜選択することができるが、液体の原料炭化水素を用いる場合は、より均一に微細に噴霧するために、供給された液を別の液体と共に噴射する2流体ノズル等、ノズルから噴霧された直後の原料炭化水素の初期液滴径が極力小さいものとするのが好ましい。原料炭化水素供給口4の開口径、形、炉内への突出具合、燃焼ガス流への供給角度、気液比などの原料炭化水素供給方法、流速、流量、温度などは、適宜選択すればよいが、第2反応帯域2に噴霧された原料炭化水素が蒸発する前に第2反応帯域2の炉壁に付着しないような条件で噴霧することが好ましい。そのように噴霧することにより、得られる煤状物質中の異物を低減することができる。

【0039】第1反応帯域1及び第2反応帯域2を構成する炉材としては、金属、耐火物など耐熱性を有する材質であれば任意のものを使用できる。金属を使用する場合は内部燃焼ガスの温度が金属の耐熱温度以上になるため、水冷ジャケット構造や水冷チューブを巻くなどの構造を採ることにより外部から冷却する必要がある。金属以外の材料としては、例えば、SiC、ダイヤモンド、窒化アルミ、窒化珪素、セラミックス系耐火材などがある。

【0040】第2反応帯域2より下流側以降は、フラレーンを含む煤状物質（反応途中のものを含む）を含んだ燃焼ガス流を1000℃以下、好ましくは800℃以下に冷却する構造とする。具体的には、反応停止流体供給口から水などを噴霧してもよいし、水冷構造等により外部を冷却した流路を通させることによって冷却を行なってもよい。特に、流路の径が小さい場合には、特に水冷構造としなくても大気への自然放熱で十分に冷却されることもある。

【0041】冷却されたフラレーン類及び煤状物質は、流路の先に設けられている捕集バグフィルター等（図示せず）でガスと分離されて回収される。フラレーン類の採取方法は、このようなバグフィルターや流路内壁に付着させる等、公知の一般的プロセスを使用することができる。

【0042】図2に示すように、本発明の第2の実施の形態に係るフラレーン類の製造装置10は、反応炉11

内に第1のバーナー12を介して供給された酸素含有ガスと燃料ガスが燃焼して高温の燃焼ガス流を形成する第1反応帯域13と、第1反応帯域13の下流側にあって、燃焼ガス流に原料炭化水素を供給する第2のバーナー14の吐出口15を有し、ガス化して供給された原料炭化水素を燃焼ガス流中で反応させてフラレーン類を生成させる第2反応帯域16を有する。以下、これらについて詳細に説明する。反応炉11は、例えば、円筒形状の側壁部17と、側壁部17の一端側に接続して徐々に外径が縮小して排出口18を形成している端部壁19とを備えている。側壁部17と端部壁19は、例えばステンレス鋼等の耐熱鋼で構成されている。更に、側壁部17の他端側の内周面には図示しない耐火物がライニングされている。耐火物としては、例えばアルミナ質の耐火煉瓦やアルミナ質の不定形耐火物を使用することができる。また、排出口18には図示しない排気管の一端側が接続され、排気管の他端側は排気ポンプに接続されている。このため、反応炉11内を大気圧未満の減圧状態にすると共に、反応炉11内で生成した煤状物質を含む燃焼ガスを反応炉11内から外部に排出することができる。

【0043】側壁部17の他端側の基盤17aに取付けられた第1のバーナー12は、酸素含有ガス供給配管20に接続した複数の酸素含有ガスノズル21と、燃料ガス供給配管22に接続した燃料ガスノズル23を有し、これらの各ガスノズル21、23は基盤17aに混在配置されている。また、酸素含有ガスノズル21、燃料ガスノズル23は、例えばステンレス鋼等の耐熱鋼で形成されている。このため、酸素含有ガスノズル21から供給された酸素含有ガスと、燃料ガスノズル23から供給された燃料ガスとは、放出された後に並流混合して一様な混合状態となって第1反応帯域13で燃焼する。そして、形成された高温の燃焼ガス流は下流側の第2反応帯域16に流入する。側壁部17の他端側に取付けられた第2のバーナー14は、第1反応帯域13を貫通して配置された多数の小径吐出管24（例えば、ステンレス鋼等の耐熱鋼で形成されている）からなっている。その結果、小径吐出管24の先端側に設けられた吐出口15は、第2反応帯域16の上流側に隙間を有して配置されている。また、各小径吐出管24の基端側は原料炭化水素供給配管25に接続している。このため、第1反応帯域13から流入する高温の燃焼ガス流中に直接原料炭化水素を均一に供給することができ、原料炭化水素を短時間に均一に熱分解することができる。

【0044】次に、本発明の第2の実施の形態に係るフラレーン類の製造装置10を使用したフラレーン類の製造方法について詳細に説明する。酸素含有ガスノズル21から酸素含有ガスを、燃料ガスノズル23から燃料ガスを供給し、これらを燃焼させることで高温の燃焼ガス流を形成し、反応炉11の下流に向かって流通させる。

酸素含有ガスとしては、酸素源である酸素ガスにアルゴンガス等の不活性ガスを任意の割合で混合したガス（例えば、不活性ガスの濃度を0、又は0を超えて90モル％以下の範囲で調整できる）を使用することができる。酸素源としては、フラーレンの収率という観点からは酸素ガスが好ましく、酸素源の入手のし易さ等の観点からは空気が好ましい。特に燃焼温度を上げるため、これらの酸素含有ガスは反応炉11内に供給される前に予熱することが好ましい。予熱の方法としては、熱交換器を使用した燃焼ガスとの熱交換、いわゆるリジェネレーションバーナ等、公知のいかなる方法を用いても良い。この予熱の温度は常温以上であればいかなる温度でも良いが、フラーレンの収率を上げるためには極力高温の方が好ましい。より好ましくは、燃焼ガスの自己着火温度以上であることが好ましい。

【0045】燃料ガスとしては、一酸化炭素、天然ガス、石油ガス等の燃料ガス、重油などの石油系液体燃料をガス化したもの、クレオソート油などの石炭系液体燃料をガス化したものを使用することができる。中でも天然ガス、石油ガス等の燃料ガスが好ましい。またフラーレンの収率を上げるためには、燃料ガスも不活性ガス等を用いて希釈することが好ましい。

【0046】続いて、燃料ガスが酸素含有ガスの下で燃焼して形成する燃焼ガス流について説明する。燃料ガスが完全燃焼する条件で燃料ガスノズル23から供給する燃料ガスの量と酸素含有ガスノズル21から供給する酸素ガスを調整して第1反応帯域13に供給すると共に、排出口18に接続された図示しない排出管を介して排気ポンプで反応炉11内を大気圧未満、より好ましくは10〜300torrの状態で保持して、図示しない着火手段で燃料ガスの燃焼を開始する。ここで、燃料ガスと酸素含有ガスは各々独立し距離を隔て分散配置された酸素含有ガスノズル21、燃料ガスノズル23から第1反応帯域13内に放出されるため、第1反応帯域13における燃焼状態を均一にすることができる。また、酸素含有ガス中の酸素ガス濃度はアルゴンガス等の不活性ガスにより希釈されて低下していることに加えて、反応炉11内の圧力が大気圧未満となっているため、第1反応帯域13での燃焼状態を高温空気燃焼状態と類似した状態にすることができる。その結果、燃料ガスの燃焼が均一に進行して、第1反応帯域13の温度を均一かつ高温（例えば、1000〜1900℃、好ましくは1700〜1900℃）にすることができる。

【0047】第2反応帯域16には、第1反応帯域13で形成された高温の燃焼ガスが流入するため、第2反応帯域16の上流側の温度は、例えば、1000〜1900℃の高温になる。原料炭化水素は、第1反応帯域13を貫通して配置された多数の小径吐出管24の各吐出口15から、第2反応帯域16の上流側の燃焼ガス流中に分散放出される。ここで、小径吐出管24は第1反応帯

域13を貫通して配置されているため、原料炭化水素は小径吐出管24内を通過中に予熱されているため、吐出口15から高温の燃焼ガス流中に放出されると直ちに熱分解する。その結果、反応活性の高い熱分解生成物が燃焼ガス中に存在し、これらが合体することによりフラーレン前駆体が形成される。そして、フラーレン前駆体が燃焼ガス流と共に移動しながら成長してフラーレンになる。なお、原料炭化水素の熱分解は吸熱反応であるため、燃焼ガスから熱エネルギーが奪われて燃焼ガスの温度が低下する。このため、原料炭化水素に酸素含有ガスを混合し、原料炭素水素の一部を燃焼させて熱エネルギーを供給するようにしてもよい。しかし、原料炭素水素の一部燃焼が活発に起こると第2反応帯域16内の温度の不均一が生じてフラーレンの生成効率が低下するため、燃焼ガス中の酸素濃度は、好ましくは3vol％以下、更に好ましくは0.05〜1vol％である。

【0048】原料炭素水素としては、従来公知の任意のものを使用することができ、例えば、ベンゼン、トルエン、キシレン、ナフタレン、アントラセン等の芳香族系炭化水素、クレオソート油、カルボン酸油などの石炭系炭化水素、エチレンヘビーエンドオイル、FCCオイル（流動接触分解残渣油）等の石油系重質油、アセチレン系不飽和炭化水素、エチレン系炭化水素、ペンタンやヘキサン等の脂肪族飽和炭化水素などが挙げられ、これらを単独又は任意の割合で混合して使用してもよい。中でも縮環した芳香族系炭化水素を用いることが好ましく、特にベンゼンやトルエン等の芳香族系炭化水素が好ましい。主に原料となる原料炭素水素の純度は高い方が好ましく、中でも芳香族系炭化水素を用いる際には純度が100％に近いほど良い。

【0049】図3に示すように、本発明の第3の実施の形態に係るフラーレン類の製造装置26は、酸素含有ガスと燃料ガスが予混合されて第1のバーナー27に供給されることが特徴である。そのため、構造が異なる第1のバーナー27についてのみ説明し、第2の実施の形態に係るフラーレン類の製造設備10と同一の構成要素には同一の符号を付して詳細な説明は省略する。第1のバーナー27は、例えば耐熱金属で作製されており、反応炉11の第1反応帯域13に一面側が露出しているヘッド28と、ヘッド28の下部に設けられた蓄圧室29を有している。そして、第2のバーナー14の各小径吐出管24は、相互に所定の隙間を開けて蓄圧室29の下方から蓄圧室29及びヘッド28を貫通し反応炉11内に突出している。

【0050】ここで、ヘッド28は、例えば焼結金属製の多孔質部材で構成されている。多孔質部材は、一面側から他面側に連通する連通孔を多数備えた構造となっており、ヘッド28の下部に設けられた蓄圧室29に酸素含有ガスと燃料ガスを予混合した混合ガスとして混合ガス供給配管30から供給すると、混合ガスはヘッド28

内の連通孔を介して蓄圧室29側の面から第1反応帯域13側に突出した面まで移動し、第1反応帯域13内に噴出することができる。従って、第1反応帯域13内に噴出した混合ガスを燃焼させることにより、第1反応帯域13で高温の燃焼ガスを形成することができる。そして、第1反応帯域13から流入する高温の燃焼ガス流中に、原料炭化水素供給配管25を介して供給した原料炭化水素を各小径吐出管24の吐出口15から供給して、原料炭化水素を短時間に均一に熱分解することができる。なお、本発明の第3の実施の形態に係るフラーレン類の製造装置26を使用したフラーレン類の製造方法は、第2の実施の形態に係るフラーレン類の製造装置10を使用したフラーレン類の製造方法と実質的に同じであるので詳細な説明は省略する。

【0051】本発明の第4の実施の形態に係るフラーレン類の製造装置31では酸素含有ガスと燃料ガスが独立に別配管で第1のバーナー32に供給されるため、第3の実施の形態に係るフラーレン類の製造装置26と第1のバーナー32の構造が異なっていることが特徴である。そのため、構造が異なる第1のバーナー32についてのみ説明し、第2の実施の形態に係るフラーレン類の製造装置10と同一の構成要素には同一の符号を付して詳細な説明は省略する。すなわち、図4に示すように、第1のバーナー32は耐熱性金属で作製され、連通孔を有する焼結金属性の多孔質部材からなるヘッド33と、ヘッド33の下部に設けられた蓄圧室34と、蓄圧室34内に噴出口を有する複数のガス混合器35を有している。そして、第2のバーナー14の各小径吐出管24は、相互に所定の隙間を開けて蓄圧室34の下から蓄圧室34及びヘッド33を貫通し反応炉11内に突出している。また、ガス混合器35としては、燃料ガスの流れて酸素含有ガスを吸引して混合するアスピレータ式の混合器を使用することができる。

【0052】このような構成とすることにより、酸素含有ガスと燃料ガスをそれぞれ独立に酸素含有ガス供給配管36及び燃料ガス供給配管37で各ガス混合器35に供給すると、酸素含有ガスと燃料ガスは混合されながらガス混合器35の噴出口から混合ガスとして蓄圧室34内に流入する。そして、蓄圧室34内に流入した混合ガスはヘッド33内の連通孔を介して蓄圧室34側の面から第1反応帯域13側に突出した面まで移動し、第1反応帯域13内に噴出することができる。従って、第1反応帯域13内に噴出した混合ガスを燃焼させることにより、第1反応帯域13で高温の燃焼ガス流を形成することができる。そして、第1反応帯域13から流入する高温の燃焼ガス流中に、原料炭化水素供給配管25を介して供給した原料炭化水素を各小径吐出管24の吐出口15から供給して、原料炭化水素を短時間に均一に熱分解することができる。

【0053】なお、本発明の第4の実施の形態に係るフ

ラーレン類の製造装置31を使用したフラーレン類の製造方法は、第3の実施の形態に係るフラーレン類の製造装置26を使用したフラーレン類の製造方法と実質的に同じであるので詳細な説明は省略する。

【0054】図5に示すように、本発明の第5の実施の形態に係るフラーレン類の製造装置38は、側壁部17の他端側の基盤17aに取付けられ、酸素含有ガスと燃料ガスが予混合された混合ガスが噴出する多数の小径の噴出ノズル39が隙間をおいて形成されているヘッダー管40を有する第1のバーナー41に供給されることが特徴である。そのため、構造が異なる第1のバーナー41についてのみ説明し、第2の実施の形態に係るフラーレンの製造装置10と同一の構成要素には同一の符号を付して詳細な説明は省略する。

【0055】ヘッダー管40は、反応炉11の軸心に対して同心上にそれぞれ隙間を設けて配置された複数の環状管40aを有し、各環状管40aは混合ガス供給配管30aに接続している。そして、第2のバーナー14の各小径吐出管24は、各環状管40aの隙間を通過して第1反応帯域13を貫通して配置されている。従って、酸素含有ガスと燃料ガスを予混合した混合ガスを混合ガス供給配管30aを介して各環状管40aに供給すると、混合ガスは各環状管40aのそれぞれの噴出ノズル39から第1反応帯域13内に噴出する。このため、第1反応帯域13内に噴出した混合ガスを燃焼させることにより、第1反応帯域13で高温の燃焼ガス流を形成することができる。そして、第1反応帯域13から流入する高温の燃焼ガス流中に、原料炭化水素供給配管25を介して供給した原料炭化水素を各小径吐出管24の吐出口15から供給して、原料炭化水素を短時間に均一に熱分解することができる。なお、本発明の第5の実施の形態に係るフラーレン類の製造装置38を使用したフラーレン類の製造方法は、第2の実施の形態に係るフラーレン類の製造装置10を使用したフラーレン類の製造方法と実質的に同じであるので詳細な説明は省略する。

【0056】本発明の第6の実施の形態に係るフラーレン類の製造装置42は、本発明の第2の実施の形態に係るフラーレン類の製造装置10と比較して、第1のバーナー43の構造が異なっていることが特徴である。そのため、構造が異なる第1のバーナー43についてのみ説明し、第2の実施の形態に係るフラーレン類の製造装置10と同一の構成要素には同一の符号を付して詳細な説明は省略する。すなわち、図6に示すように、側壁部17の他端側の基盤17aに取付けられた第1のバーナー43は耐熱性金属で作製され、酸素含有ガスを噴出する多数の小径の噴出ノズル44が隙間をおいて形成された第1のヘッダー管45と、第1のヘッダー管45とは隙間を有し配置され燃料ガスを噴出する多数の小径の噴出ノズル46が隙間をおいて形成された第2のヘッダー管47を有している。更に、第1のヘッダー管45及び第

2のヘッダー管47には酸素含有ガス及び前記燃料ガスをそれぞれ独立に供給する酸素含有ガス供給配管20、燃料ガス供給配管22が接続されている。また、第2のバーナー14の小径吐出管24は、第1のヘッダー管45と第2のヘッダー管47の間隙を通過して基盤17aを貫通し反応炉11内に突出している。

【0057】このような構成とすることにより、酸素含有ガスを酸素含有ガス供給配管20を介して第1のヘッダー管45に供給し噴出ノズル44から反応炉11内に噴出させることができる。また、燃料ガスを燃料ガス供給配管22を介して第2のヘッダー管47に供給し噴出ノズル46から反応炉11内に噴出させることができる。各噴出ノズル44、46から噴出した酸素含有ガスと燃料ガスとは、放出された後に並流混合して一様な混合状態となって第1反応帯域13で燃焼する。そして、形成された高温の燃焼ガスは下流側の第2反応帯域16に流入する。そして、第1反応帯域13から流入する高温の燃焼ガス流中に、原料炭化水素供給配管25を介して供給した原料炭化水素を各小径吐出管24の吐出口15から供給して、原料炭化水素を短時間に均一に熱分解することができ、なお、本発明の第6の実施の形態に係るフラーレン類の製造装置42を使用したフラーレン類の製造方法は、第2の実施の形態に係るフラーレン類の製造装置10を使用したフラーレン類の製造方法と実質的に同じであるので詳細な説明は省略する。

【0058】以上、本発明の実施の形態を説明したが、本発明は、この実施の形態に限定されるものではなく、発明の要旨を変更しない範囲での変更は可能であり、前記したそれぞれの実施の形態や変形例の一部又は全部を組み合わせて本発明のフラーレン類の製造方法及びその装置を構成する場合も本発明の権利範囲である。例えば、第5の実施の形態でヘッダー管40を反応炉11の軸心に対して同心上に配置された複数の環状管40aで構成したが、複数の直管を格子状にそれぞれ隙間を設けて並べてもよい。また、第6の実施の形態で第1のヘッダー管45と第2のヘッダー管47を反応炉11の軸心に対して同心上に隙間を設けて複数配置したが第1のヘッダー管と第2のヘッダー管を格子状にそれぞれ隙間を設けて並べてもよい。更に、第2のバーナー14の小径吐出管24をステンレス鋼等の耐熱鋼で作製し、第3及び第4の実施の形態で多孔質部材を耐熱性の焼結金属で作製したが、サーメット、セラミックスで作製することもできる。

【0059】

【発明の効果】請求項1～3記載のフラーレン類の製造方法においては、反応炉内に、酸素含有ガスと燃料とを供給して燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、この燃焼ガス流の途中にガス化した原料炭化水素を供給する原料炭化水素供給口を有し且つ原料炭化水素を反応させてフラーレン類を生成させる第2反応

帯域を有することを特徴とするフラーレン類の製造装置を使用し、第2反応帯域の圧力を大気圧未満とするので、原料炭化水素の熱分解が均一に進行して、フラーレン類の生成効率を向上させることができ、フラーレン類を大量に且つ安価に、そして容易に製造することができる。

【0060】一方、上記、公知の燃焼法によるフラーレン類の製造方法においては、燃焼反応のための燃料とフラーレン生成のための原料は同一であるのが通常であり、炭化水素燃料燃焼反応に必要な燃料を任意に選定することができない。これに対し、本発明によると燃焼反応のための燃料と、フラーレン類の製造のための原料を別々に選定することができるため、特に工業規模でフラーレン類を製造する場合、原料の調達率倍により、コストの安い原料を自由に選択することができる。

【0061】特に、請求項2記載のフラーレン類の製造方法においては、第2反応帯域が第1反応帯域の下流側にあるので、第2反応帯域の条件を炉内断面すべてにわたって一定に保つことができ、この帯域での条件をフラーレン類の収率が最大となるような条件に調節することによって、フラーレン類が生成する領域を最大に広げることができるため、通常の燃焼法に比べてフラーレン類の収率が高くなる。これに対して、従来の燃焼法においては主に火炎中でフラーレン類が生成するが、一時的に火炎は温度分布を持ち、火炎の特定の領域でフラーレン類が生成することが知られている。

【0062】請求項3記載のフラーレン類の製造方法においては、第2反応帯域の温度が1000℃以上であるので、供給された原料炭化水素を短時間に確実に熱分解させることができ、フラーレン類を大量に製造することができる。

【0063】請求項4～13記載のフラーレン類の製造装置においては、反応炉内に、第1のバーナーを介して酸素含有ガスと燃料ガスを供給し、これらを燃焼させて高温の燃焼ガス流を形成させる第1反応帯域と、第1反応帯域の下流側にあって、燃焼ガス流に原料炭化水素を供給する第2のバーナーの吐出口を有し、ガス化して供給された原料炭化水素を燃焼ガス流中で反応させてフラーレン類を生成させる第2反応帯域を有するので、燃料の燃焼状態の制御、原料炭化水素の熱分解の制御が共に容易となって、フラーレン類を大量に、安価に、そして容易に製造することが可能となる。

【0064】特に、請求項5記載のフラーレン類の製造装置においては、第2のバーナーの吐出口は、第2反応帯域の上流側に隙間を有して多数形成され、原料炭化水素を燃焼ガス流中に分散放出するので、燃焼ガス中で原料炭化水素を短時間に均一に熱分解することができ、原料炭化水素の熱分解物から生成させるフラーレン類の収率を高くすることが可能となる。

【0065】請求項6記載のフラーレン類の製造装置に

においては、第2のバーナーは、第1反応帯域を貫通して配置される多数の小径吐出管からなっている。第2反応帯域の高温の燃焼ガス流中に予熱された原料炭化水素を一樣に分散放出して熱分解することができ、原料炭化水素の熱分解物から生成させるフラレーン類の収率を高くすることが可能となる。

【0066】請求項7記載のフラレーン類の製造装置においては、第1のバーナーは、酸素含有ガスと燃料ガスとを独立に放出し混合配置された複数の酸素含有ガスノズル及び燃料ガスノズルを有するので、供給された酸素含有ガスと燃料ガスは拡散混合して一様な混合状態で第1反応帯域に存在させることができ、燃料ガスを第1反応帯域で容易に完全燃焼させることが可能となる。その結果、高温の燃焼ガス流を形成することができ、原料炭化水素の熱分解物から生成させるフラレーン類の収率を高くすることが可能となる。

【0067】請求項8記載のフラレーン類の製造装置においては、第1のバーナーのヘッドは多孔質部材からなっており、表面から酸素含有ガスと燃料ガスが混合された状態で噴出されるので、酸素含有ガスと燃料ガスを予混合された状態で第1反応帯域に供給することができ、燃料ガスを第1反応帯域で容易に完全燃焼させることが可能となる。その結果、高温の燃焼ガス流を形成することができ、原料炭化水素の熱分解物から生成させるフラレーン類の収率を高くすることが可能となる。

【0068】請求項9記載のフラレーン類の製造装置においては、酸素含有ガスと燃料ガスの混合は第1のバーナー内で行われ、第1のバーナーには酸素含有ガスと燃料ガスが独立に別配管で供給されているので、酸素含有ガスと燃料ガスとの予混合手段を設ける必要がなく、フラレーン類の製造装置の構成を簡単にすることができる。

【0069】請求項10記載のフラレーン類の製造装置においては、酸素含有ガスと燃料ガスとは予混合されてヘッドの下部に設けられた蓄圧室に供給されているので、第1のバーナーの構造を簡単にすることができ、第1のバーナーのコストを低減させることができる。

【0070】請求項11記載のフラレーン類の製造装置においては、第1のバーナーは、多数の小径の噴出ノズルが隙間を有して形成されたヘッダー管を有し、ヘッダー管には予混合された酸素含有ガスと燃料ガスが供給されているので、酸素含有ガスと燃料ガスを予混合された状態で第1反応帯域に分散放出することができ、燃料ガスを第1反応帯域で容易に完全燃焼させることが可能となる。その結果、高温の燃焼ガス流を形成することができ、原料炭化水素の熱分解物から生成させるフラレーン類の収率を高くすることが可能となる。

【0071】請求項12記載のフラレーン類の製造装置においては、第1のバーナーは、酸素含有ガスを噴出する多数の小径の噴出ノズルが隙間を有して形成された第

1のヘッダー管と、第1のヘッダー管とは隙間を有し配置され燃料ガスを噴出する多数の小径の噴出ノズルが隙間を有して形成された第2のヘッダー管を有し、第1のヘッダー管及び第2のヘッダー管には酸素含有ガス及び燃料ガスがそれぞれ独立に別配管で供給されているので、分散放出された酸素含有ガスと燃料ガスは拡散混合して一様な混合状態となって第1反応帯域に存在させることができ、燃料ガスを第1反応帯域で容易に完全燃焼させることが可能となる。その結果、高温の燃焼ガス流を形成することができ、原料炭化水素の熱分解物から生成させるフラレーン類の収率を高くすることが可能となる。

【0072】請求項13記載のフラレーン類の製造装置においては、第2のバーナーから供給される原料炭化水素に酸素含有ガスを混合するので、原料炭化水素が熱分解する際に消費した熱エネルギーを補填して燃焼ガスの温度が低下するのを防止でき、原料炭化水素の熱分解物から生成させるフラレーン類の収率を高くすることが可能となる。

【図面の簡単な説明】

【図1】(A)、(B)はそれぞれ本発明の第1の実施の形態に係るフラレーン類の製造方法を適用したフラレーン製造装置の説明図、平断面図である。

【図2】(A)、(B)はそれぞれ本発明の第2の実施の形態に係るフラレーン類の製造装置の説明図、平断面図である。

【図3】(A)、(B)はそれぞれ本発明の第3の実施の形態に係るフラレーン類の製造装置の説明図、平断面図である。

【図4】本発明の第4の実施の形態に係るフラレーン類の製造装置の部分説明図である。

【図5】(A)、(B)はそれぞれ本発明の第5の実施の形態に係るフラレーン類の製造装置の説明図、平断面図である。

【図6】(A)、(B)はそれぞれ本発明の第6の実施の形態に係るフラレーン類の製造装置の説明図、平断面図である。

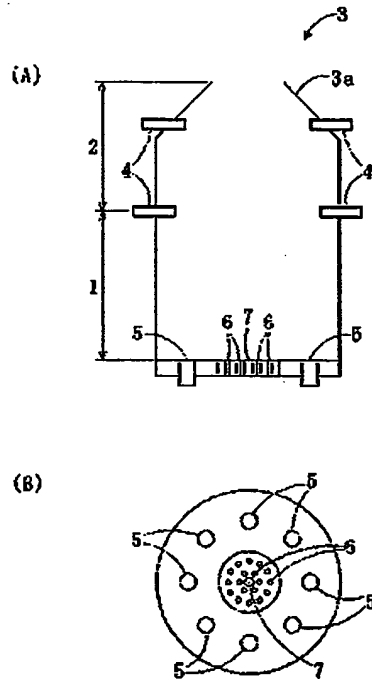
【符号の説明】

1：第1反応帯域、2：第2反応帯域、3：フラレーン類の製造装置、3a：反応炉、4：原料炭化水素供給口、5、6：酸素含有ガス供給口、7：燃料供給口、10：フラレーン類の製造装置、11：反応炉、12：第1のバーナー、13：第1反応帯域、14：第2のバーナー、15：吐出口、16：第2反応帯域、17：側壁部、17a：基盤、18：排出口、19：燃焼室、20：酸素含有ガス供給配管、21：酸素含有ガスノズル、22：燃料ガス供給配管、23：燃料ガスノズル、24：小径吐出管、25：原料炭化水素供給配管、26：フラレーン類の製造装置、27：第1のバーナー、28：ヘッド、29：蓄圧室、30、30a：混合ガス

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供給配管、31：フラーレン製の製造装置、32：第1のバーナー、33：ヘッド、34：蓄圧室、35：ガス混合器、36：酸素含有ガス供給配管、37：燃料ガス供給配管、38：フラーレン製の製造装置、39：噴出\*

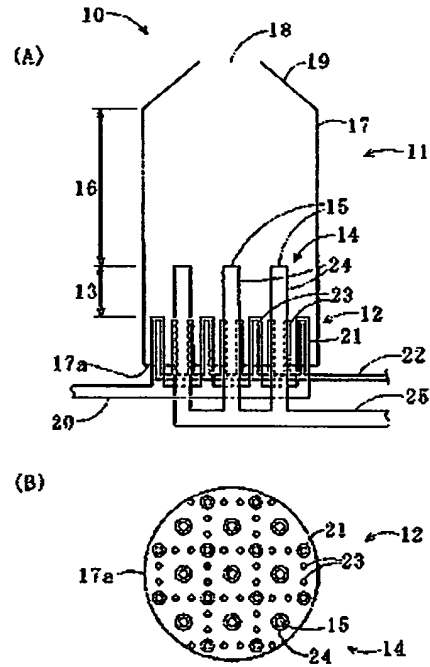
【図1】



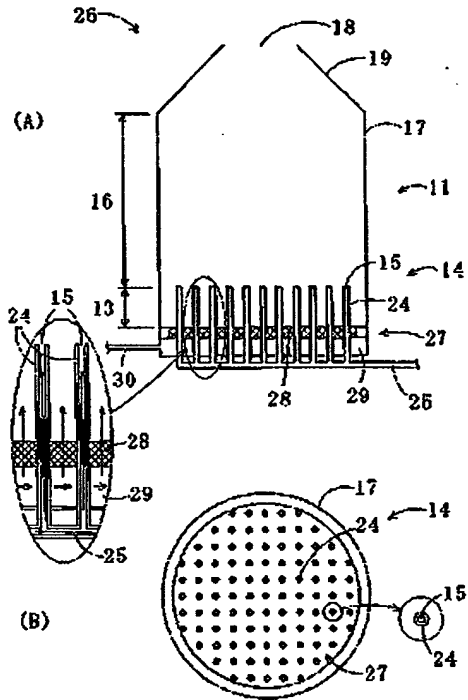
22

\*ノズル、40：ヘッダー管、40a：環状管、41：第1のバーナー、42：フラーレン製の製造装置、43：第1のバーナー、44：噴出ノズル、45：第1のヘッダー管、46：噴出ノズル、47：第2のヘッダー管

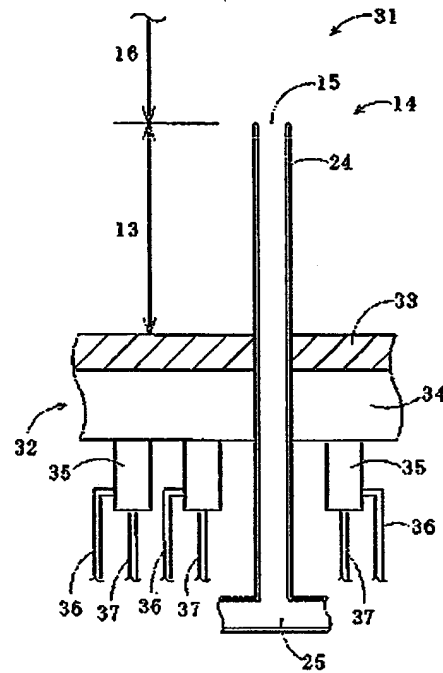
【図2】



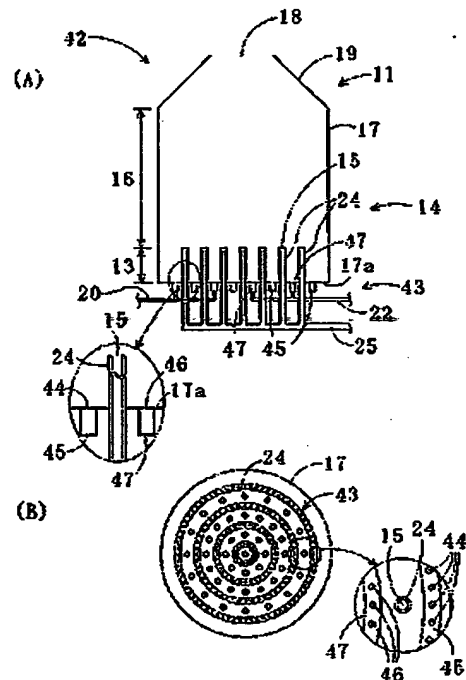
【図3】



【図4】

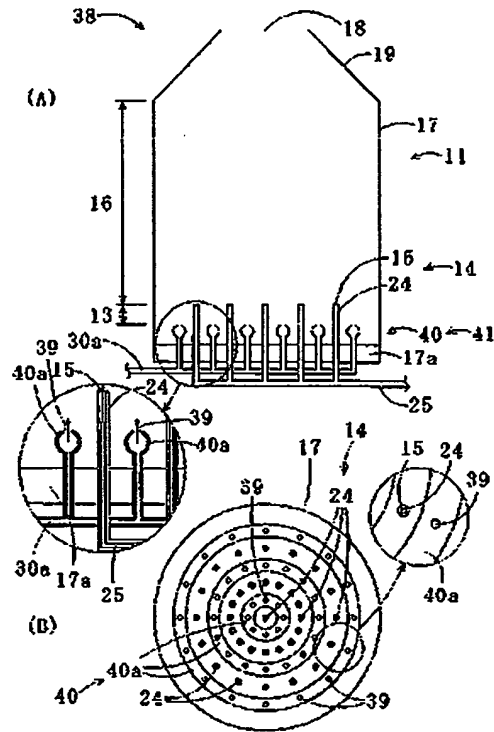


【図6】





【図5】



フロントページの続き

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Fターム(参考) 4G146 AA07 BA12 BC03 BC07 BC08  
BC27 BC34A BC34B BC35A  
BC35B BC36A BC38A BC38B  
DA03 DA23 DA25 DA35

**\* NOTICES \***

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**CLAIMS**

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[Claim(s)]

[Claim 1] The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the middle of this combustion gas style, and having the 2nd reaction band which makes this coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. The manufacture approach of the fullerene characterized by making the pressure of said 2nd reaction band under into atmospheric pressure.

[Claim 2] The manufacture approach of the fullerene characterized by said 2nd reaction band being in the downstream of said 1st reaction band in the manufacture approach of fullerene according to claim 1.

[Claim 3] The manufacture approach of the fullerene characterized by the temperature of said 2nd reaction band being 1000 degrees C or more in the manufacture approach of fullerene given in any 1 term of claims 1 and 2.

[Claim 4] The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, Are in the downstream of this 1st reaction band, and it has the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen in the style of [ said ] combustion gas. The manufacturing installation of the fullerene characterized by having the 2nd reaction band which makes said coal-for-coke-making-ized hydrogen gasified and supplied react in said combustion gas style, and makes fullerene generate.

[Claim 5] It is the manufacturing installation of the fullerene which the delivery of said 2nd burner has a clearance in the upstream of said 2nd reaction band in the manufacturing installation of fullerene according to claim 4, and are characterized by carrying out a large number formation and carrying out distributed emission of said coal-for-coke-making-ized hydrogen into said combustion gas style.

[Claim 6] It is the manufacturing installation of the fullerene characterized by consisting of a minor diameter discharge tube of a large number arranged by said 2nd burner penetrating said 1st reaction band in the manufacturing installation of fullerene according to claim 5.

[Claim 7] It is the manufacturing installation of the fullerene characterized by carrying out mixture arrangement of two or more oxygen content gas nozzles and fuel gas nozzles to which said 1st burner emits independently said oxygen content gas and said fuel gas in the manufacturing installation of fullerene given in any 1 term of claims 4-6, respectively.

[Claim 8] It is the manufacturing installation of the fullerene characterized by for the head of said 1st burner consisting of a porosity member in the manufacturing installation of fullerene given in any 1 term of claims 4-6, and blowing off where said oxygen content gas and said fuel gas are mixed from a front face.

[Claim 9] It is the manufacturing installation of the fullerene characterized by performing mixing of said oxygen content gas and said fuel gas within said 1st burner, and supplying independently said oxygen content gas and said fuel gas to said 1st burner for another piping in the manufacturing installation of fullerene according to claim 8.

[Claim 10] It is the manufacturing installation of the fullerene characterized by supplying the accumulator which premixing of said oxygen content gas and said fuel gas was carried out in the manufacturing installation of fullerene according to claim 8, and was prepared in the lower part of said head.

[Claim 11] Said 1st burner is the manufacturing installation of the fullerene which have header tubing with which the jet nozzle of many minor diameters set the clearance in the manufacturing installation of fullerene given in any 1 term of claims 4-6, and was formed, and are characterized by supplying said oxygen content gas with which premixing was carried out to this header tubing, and said fuel gas.

[Claim 12] In the manufacturing installation of fullerene given in any 1 term of claims 4-6 said 1st burner The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout said oxygen content gas set the clearance, and was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with said 1st header tubing, are arranged, and spout said fuel gas set the clearance, and was formed. The manufacturing installation of the fullerene characterized by supplying independently said oxygen content gas and said fuel gas to said 1st header tubing and said 2nd header tubing for another piping, respectively.

[Claim 13] The manufacturing installation of the fullerene characterized by mixing oxygen content gas in the coal-for-coke-making-ized hydrogen supplied from said 2nd burner in the manufacturing installation of fullerene given in any 1 term of claims 4-12.

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[Translation done.]

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacture approach of fullerene, and its equipment.

[0002]

[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings so that it might be represented by C60 and C70 grade. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
- (3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below is mentioned as the manufacture approach of fullerene learned now.

- (1) It is the approach of irradiating the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, and compounding by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot of laser is slight and it is unsuitable for extensive manufacture.
- (2) It is the approach to which carry out energization heating and a graphite rod is made to sublimate in the container under the reduced pressure filled with resistance heating method gaseous helium. Since the electric resistance loss in a circuit is large, it is unsuitable for extensive manufacture.

[0004] (3) It is the approach to which the carbon of a lifting and an anode plate is made to sublimate arc

discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in number of arc discharge methods 10kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in a current works scale.

(4) Instead of using radio frequency heating method resistance heating and arc discharge, it is the approach of heating an eddy current to raw material graphite by RF induction, and evaporating a sink and raw material graphite.

(5) It is the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. It is observed as the mass-producing method for being a point usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc discharge method in the soot (fullerene etc.) which sub\*\* in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.

(6) It is the approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0005] Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene in large quantities cheaply by any approach until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, for example, the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the patent reference 1, and collecting condensates in it is indicated. This approach is an approach of manufacturing fullerene by burning a carbon inclusion in a flame, and the fuel for combustion and the raw material of fullerene are the same carbon inclusions substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black.

[0006] As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is industrially mentioned as the general manufacture approach. The carbon black manufacturing installation (fission reactor) of the shape for example, of a cylinder is used for this approach. To \*\*\*\*, to horizontal or a perpendicular direction, supply oxygen content gas and fuels, such as air, and they are burned in the 1st reaction band of the fission reactor concerned. It is made to move to the 2nd reaction band with the cross section which was installed in the lower stream of a river of furnace shaft orientations, and reduced the obtained combustion gas style. It is the approach of supplying coal-for-coke-making-sized hydrogen (stock oil), making it reacting into the gas stream concerned, making carbon black generating, quenching gas by spraying of cooling water etc. to a gas stream further in the 3rd reaction band on the lower stream of a river, and stopping a reaction.

[0007]

[Patent reference 1] \*\*\*\*\* No. 507879 [ six to ] official report [0008]

[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the above-mentioned usual carbon black, fullerene is hardly generated. In manufacture of fullerene, it has been a big technical problem how the rate of the fullerene contained in the soot-like matter obtained is raised. Generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [ these reduced pressure ] that diluent concentration will affect the yield of the above-mentioned fullerene.

[0009] In order to raise the yield of fullerene in the above and the patent reference 1, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating which heats a flame directly, microwave heating, discharge heating, and counterflow heating that heats a flame by heat exchange with elevated-temperature gas are mentioned.

[0010] By the above and the patent reference 1, pure oxygen is used as an oxidizer for a combustion reaction, and the argon is used as a diluent. This is considered to be effective in gathering the yield of fullerene. However, the amount of the oxygen needed for combustion also becomes extensive, and pure oxygen becomes a special oxygen supply facility is required and expensive [ the manufacturing cost of

fullerene ] as a result, when a bomb or supply equipment of dedication etc. tends to be required and it is going to manufacture fullerene on a scale of industry especially.

[0011] So, it has not result in utilization for the reasons of the rate which the volume increases at the time of the actuation under that combustion temperature becomes low since there are many rates of that a flame is not stabilize compared with pure oxygen since the oxygen density is low although it can guess easily use air as an oxidizer of combustion in order to reduce a manufacturing cost in a combustion method, or nitrogen, especially reduced pressure, and passes a nozzle become quick. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the technique of manufacturing fullerene cheaply and easily in large quantities is desired.

[0012] This invention is made in view of a situation which was mentioned above, and it aims at offering the manufacture approach of the fullerene which manufacture fullerene cheaply and easily in large quantities, and its equipment.

[0013]

[Means for Solving the Problem] The first reaction band in which this invention persons supply oxygen content gas and a fuel, and burn them in a fission reactor as a result of examining various the optimal combustion methods and manufacturing installations which can manufacture fullerene in large quantities and cheaply, and a hot combustion gas style is made to form, The manufacturing installation of the fullerene which have the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and have the 2nd reaction band which makes coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. Knowledge that fullerene is stably generable in large quantities by maintaining the pressure of the 2nd reaction band under at atmospheric pressure was acquired.

[0014] Namely, the manufacture approach of the fullerene concerning the 1st invention in alignment with said purpose The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the middle of this combustion gas style, and having the 2nd reaction band which makes this coal-for-coke-making-ized hydrogen react and makes fullerene generate is used, and the pressure of said 2nd reaction band is made under into atmospheric pressure. Since a fuel and oxygen content gas are supplied and are burned in the 1st reaction band, perfect combustion can be attained easily, for example and a hot combustion gas style can be formed. And by supplying coal-for-coke-making-ized hydrogen into the acquired hot gas stream, the pyrolysis of the coal-for-coke-making-ized hydrogen can be carried out easily, and the generation effectiveness of fullerene can be raised. Moreover, by making the pressure in the 2nd reaction band under into atmospheric pressure, and rarefying the mixed state of coal-for-coke-making-ized hydrogen and combustion gas, the pyrolysis of coal-for-coke-making-ized hydrogen can advance to homogeneity and the generation effectiveness of fullerene can be raised.

[0015] In the manufacture approach of the fullerene concerning the 1st invention, it is desirable that said 2nd reaction band is in the downstream of said 1st reaction band. By establishing the 2nd reaction band in the downstream of the 1st reaction band, the hot combustion gas formed in the 1st reaction band can be immediately introduced into the 2nd reaction band. Consequently, temperature of the 2nd reaction band can be made into an elevated temperature. In the manufacture approach of the fullerene concerning the 1st invention, it is desirable that the temperature of said 2nd reaction band is 1000 degrees C or more. By making temperature of the 2nd reaction band into 1000 degrees C or more, the pyrolysis of the supplied coal-for-coke-making-ized hydrogen can be carried out in a short time certainly.

[0016] The manufacturing installation of the fullerene concerning the 2nd invention in alignment with said purpose The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, It is in the downstream of this 1st reaction band, and has the 2nd reaction band which makes said coal-for-coke-making-ized hydrogen which has the delivery of the 2nd burner which supplies coal-for-coke-making-

ized hydrogen in the style of [ said ] combustion gas, and was gasified and supplied react in said combustion gas style, and makes fullerene generate. Since combustion of a fuel is performed in the 1st reaction band, control of a combustion condition becomes easy and hot combustion gas can be formed easily. Control of the pyrolysis of coal-for-coke-making-ized hydrogen becomes easy by introducing the obtained hot combustion gas style into the 2nd reaction band, and adjusting gas stream conditions, such as temperature of a hot combustion gas style, the rate of flow, and a flow rate, and the conditions of supply of coal-for-coke-making-ized hydrogen, since the pyrolysis of the coal-for-coke-making-ized hydrogen is supplied and carried out into this hot gas stream.

[0017] As for the delivery of said 2nd burner, in the manufacturing installation of the fullerene concerning the 2nd invention, it is desirable to have a clearance in the upstream of said 2nd reaction band, and for a large number formation to be carried out and to carry out distributed emission of said coal-for-coke-making-ized hydrogen into said combustion gas style. By forming in the upstream of the 2nd reaction band the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen, direct coal-for-coke-making-ized hydrogen can be supplied into the hot combustion gas style which flows from the 1st reaction band, and the pyrolysis of the coal-for-coke-making-ized hydrogen can be carried out easily. Moreover, since distributed emission of the coal-for-coke-making-ized hydrogen is carried out into combustion gas from many deliveries, coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in combustion gas in a short time. As for said 2nd burner, in the manufacturing installation of the fullerene concerning the 2nd invention, it is desirable to consist of a minor diameter discharge tube of a large number arranged by penetrating said 1st reaction band. Since coal-for-coke-making-ized hydrogen is supplied with many minor diameter discharge tubes, distributed emission of the coal-for-coke-making-ized hydrogen can be uniformly carried out into the combustion gas style of the elevated temperature of the 2nd reaction band. Moreover, since a minor diameter discharge tube penetrates the 1st reaction band and is arranged, coal-for-coke-making-ized hydrogen is gradually heated by hot combustion gas, passing through the inside of a minor diameter discharge tube, and can promote the pyrolysis in the inside of the combustion gas style of the elevated temperature of the 2nd reaction band.

[0018] In the manufacturing installation of the fullerene concerning the 2nd invention, mixture arrangement of two or more oxygen content gas nozzles and fuel gas nozzles to which said 1st burner emits independently said oxygen content gas and said fuel gas, respectively may be carried out. Diffusive mixing of the oxygen content gas and fuel gas which were supplied can be carried out, they can be in the uniform mixed state, and can be made to exist in the 1st reaction band by considering as such a configuration. Moreover, in the manufacturing installation of the fullerene concerning the 2nd invention, the head of said 1st burner consists of a porosity member, and can be considered as the configuration which blows off from a front face where said oxygen content gas and said fuel gas are mixed. By considering as such a configuration, oxygen content gas and fuel gas can be supplied to the 1st reaction band, where premixing is carried out.

[0019] In the manufacturing installation of the fullerene concerning the 2nd invention, mixing of said oxygen content gas and said fuel gas is performed within said 1st burner, and it can consider as the configuration to which said oxygen content gas and said fuel gas are independently supplied for another piping at said 1st burner. Since mixing of oxygen content gas and fuel gas is performed within the 1st burner, it is not necessary to establish separately the premixing means of oxygen content gas and fuel gas, and the configuration of the manufacturing installation of fullerene becomes easy. In the manufacturing installation of the fullerene concerning the 2nd invention, said oxygen content gas and said fuel gas can be considered as the configuration supplied to the accumulator which premixing was carried out and was prepared in the lower part of said head. Since premixing of oxygen content gas and the fuel gas is carried out and they are supplied to the accumulator of the lower part of a head, structure of the 1st burner can be simplified.

[0020] In the manufacturing installation of the fullerene concerning the 2nd invention, said 1st burner has header tubing with which the jet nozzle of many minor diameters set the clearance, and was formed, and can consider it as the configuration to which said oxygen content gas with which premixing was

carried out to this header tubing, and said fuel gas are supplied. By considering as such a configuration, where premixing is carried out, distributed emission of oxygen content gas and the fuel gas can be carried out in the 1st reaction band. In the manufacturing installation of the fullerene concerning the 2nd invention said 1st burner The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout said oxygen content gas set the clearance, and was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with said 1st header tubing, are arranged, and spout said fuel gas set the clearance, and was formed. It can consider as the configuration to which said oxygen content gas and said fuel gas are independently supplied for another piping, respectively at said 1st header tubing and said 2nd header tubing. Diffusive mixing of the oxygen content gas and fuel gas by which distributed emission was carried out can be carried out, they can be in the uniform mixed state, and can be made to exist in the 1st reaction band by considering as such a configuration.

[0021] In the manufacturing installation of the fullerene concerning the 2nd invention, oxygen content gas is mixable in the coal-for-coke-making-ized hydrogen supplied from said 2nd burner. In the pyrolysis of coal-for-coke-making-ized hydrogen, the temperature of combustion gas falls by the pyrolysis of coal-for-coke-making-ized hydrogen for endothermic reaction. For this reason, it can prevent that fill up the heat energy consumed when a part of coal-for-coke-making-ized hydrogen was burned in the 2nd reaction band, heat energy was generated and coal-for-coke-making-ized hydrogen pyrolyzed by mixing oxygen content gas in coal-for-coke-making-ized hydrogen, and the temperature of combustion gas falls.

[0022]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented. The explanatory view of the fullerene manufacturing installation which applied the manufacture approach of the fullerene which drawing 1 (A) and (B) require for the gestalt of operation of the 1st of this invention, respectively here, The explanatory view of the manufacturing installation of the fullerene which a plane section Fig., drawing 2 (A), and (B) require for the gestalt of operation of the 2nd of this invention, respectively, The explanatory view of the manufacturing installation of the fullerene which a plane section Fig., drawing 3 (A), and (B) require for the gestalt of operation of the 3rd of this invention, respectively, A plane section Fig., the partial explanatory view of the manufacturing installation of the fullerene which drawing 4 requires for the gestalt of operation of the 4th of this invention, The explanatory view of the manufacturing installation of the fullerene which drawing 5 (A) and (B) require for the gestalt of operation of the 5th of this invention, respectively, a plane section Fig., drawing 6 (A), and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 6th of this invention, and a plane section Fig., respectively.

[0023] The manufacture approach of the fullerene concerning the gestalt of operation of the 1st of this invention is explained using drawing 1. The manufacture approach of the fullerene concerning the gestalt of the 1st operation is related with the approach of introducing coal-for-coke-making-ized hydrogen into the manufacturing installation 3 of the fullerene constituted by forming the 1st reaction band 1 and the 2nd reaction band 2 in fission reactor 3a, and manufacturing fullerene by burning.

[0024] The manufacturing installation 3 of fullerene has the 2nd reaction band 2 which coal-for-coke-making-ized hydrogen is supplied [ band ], makes it react the 1st reaction band 1 in which a combustion gas style is made to form, and in the style of [ which were formed there ] combustion gas, and makes fullerene generate. The 2nd reaction band 2 may be in the downstream of the direction of a combustion gas style (it may be hereafter called "shaft orientations") which may be the almost same field (an outside or inside) as the 1st reaction band 1, and was formed in the 1st reaction band 1.

[0025] Drawing 1 shows the case where the 2nd reaction band 2 is located on the lower stream of a river of the 1st reaction band 1.

Generally in the [1st reaction band] 1st reaction band 1, a combustion gas style hot by supplying a fuel and oxygen content gas and burning them, respectively is generated toward the lower stream of a river of the 2nd reaction band 2, i.e., fission reactor 3a, from a fuel feed hopper and oxygen content gas



supply opening.

[0026] Even if supply of a fuel and oxygen content gas is the so-called premixing method mixed before entering in fission reactor 3a, it may be the so-called diffusive-mixing method supplied to fission reactor 3a from the nozzle which became independent, respectively. In drawing 1, in the case of a diffusive-mixing method, a fuel is supplied from the central fuel feed hopper 7, and it supplies oxygen content gas from the oxygen content gas supply openings 5 and 6 of the perimeter. Moreover, a premixing method and a diffusive-mixing method may be combined, for example, in drawing 1, from the oxygen content gas supply opening 5, what mixed oxygen content gas with the fuel beforehand may be supplied, and the fuel from the fuel feed hopper 7 may be independently supplied for oxygen content gas from the oxygen content gas supply opening 6, respectively.

[0027] It may be the purpose that this 1st reaction band 1 generates hot combustion gas, and that combustion method may be what kind of well-known combustion methods, such as premixed combustion, diffusive burning, laminar-flow combustion, turbulent flow combustion, and elevated-temperature air combustion. Moreover, although combustion in the 1st reaction band 1 may be perfect combustion or you may be incomplete combustion as long as the temperature which becomes generable [ fullerene ] in the 2nd reaction band 2 is acquired, it is desirable that it is perfect combustion with the large calorific value to fuel used. When the 1st reaction band 1 is incomplete combustion with the so-called superfluous fuel, the soot-like matter which contains fullerene even in the 1st reaction band 1 may generate.

[0028] However, the combustion by the lean mixture whose oxygen required for combustion is more than the amount of stoichiometry oxygen of the combustion in this 1st reaction band 1 is preferably better. As oxygen content gas, the gas which mixed non-flammable gas, such as argon gas and nitrogen gas, at a rate of arbitration can be used for air, oxygen gas, or these. NOX especially in elevated-temperature combustion Pure oxygen may be used in order to suppress generating. In order to gather the yield of fullerene, it is desirable to dilute using rare gas etc. in a combustion process. Rare gas may be supplied from the exclusive nozzle for supply, and may be beforehand mixed in a fuel, coal-for-coke-making-ized hydrogen, and oxygen content gas.

[0029] As a fuel, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, a fuel oil, benzene, and toluene, and creosote oil, can be used. Especially, as a fuel used with the gestalt of this operation, fuel gas is desirable. Moreover, although what is necessary is for fullerene to obtain just to adjust suitably the mean temperature in the 1st reaction band 1 at the time of fullerene manufacture, it is preferably made into 1600 degrees C or more still more preferably 1300 degrees C or more. This is because the productivity of fullerene goes up, so that the temperature of combustion gas is an elevated temperature. Even if an upper limit is too high not much, the productivity of fullerene may fall. Moreover, what is necessary is just to determine after taking into consideration the heat-resistant problem by the quality of the material of a fission reactor.

[0030] If opening of the arrangement of the fuel feed hopper 7 and the oxygen content gas supply openings 5 and 6 is carried out to fission reactor 3a, it is arbitrary. In drawing 1, opening of the fuel feed hopper 7 and the oxygen content gas supply openings 5 and 6 is carried out to the same fission reactor 3a side. The configuration of each feed hoppers 5, 6, and 7 which are carrying out opening into fission reactor 3a may be arbitrary, and may be the indeterminate form of the shape of a polygon, such as an approximate circle form, an ellipse form, and the shape of a trigonum and a rectangular head, a gourd mold, etc.

[0031] As for fission reactor 3a internal pressure, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

Coal-for-coke-making-ized hydrogen is supplied from the coal-for-coke-making-ized hydrogen feed hopper 4 in the style of [ which was formed in the 1st reaction band 1 ] combustion gas, and fullerene is made to generate in the [2nd reaction band] 2nd reaction band 2 by carrying out partial combustion of a part of this coal-for-coke-making-ized hydrogen. In order to carry out partial combustion, it is good also considering the combustion in the 1st reaction band 1 as hyperoxia so that oxygen may remain.

Moreover, a nozzle may be arranged to the 2nd reaction field 2, and oxygen content gas may be supplied to it from an oxygen content gas supply nozzle.

[0032] Under the present circumstances, as for the above-mentioned coal-for-coke-making-ized hydrogen supplied into combustion gas, or oxygen content gas, it is desirable to be supplied in fission reactor 3a as much as possible at homogeneity. For this reason, it is desirable to be equally arranged so well that many by the number of the coal-for-coke-making-ized hydrogen feed hopper 4 installed in the 2nd reaction band 2 and an oxygen content gas supply nozzle in fission reactor 3a.

[0033] What is necessary is just to choose the die length of the 2nd reaction band suitably according to the magnitude of fission reactor 3a, the class of fullerene to manufacture, etc. The location and configuration of the 2nd reaction band may be arbitrary, and may be the inside of the 1st reaction band, or may be an outside, and as shown in drawing 1, they may be in the downstream of the 1st reaction band 1. It is more desirable for the cross-section configuration of the 2nd reaction band not to change, although the configuration of the 2nd reaction band is also arbitrary. The reason is that it will have effect which is not desirable on the fullerene to generate if influenced by the flow by the cross-section configuration of the 2nd reaction band changing in the process which fullerene generates of turbulence.

[0034] Although what is necessary is just to choose the mean temperature of the 2nd reaction band 2 suitably by the fullerene to manufacture, in order that coal-for-coke-making-ized hydrogen may evaporate and react to homogeneity, it is desirable that it is an elevated-temperature ambient atmosphere enough. It is desirable that it is specifically 1000 degrees C or more, and it is especially desirable that it is 1700-1900 degrees C especially. Moreover, in the 2nd reaction band 2, it is desirable to control the oxygen density in combustion gas as much as possible. It is because there is a thing of coal-for-coke-making-ized hydrogen, the generation reaction band 2, i.e., 2nd reaction band, of fullerene, which combustion takes place actively in part, therefore the ununiformity of the temperature in the 2nd reaction band 2 produces when oxygen exists so much in combustion gas. the oxygen density in combustion gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably.

[0035] In the gestalt of this operation, the location which supplies coal-for-coke-making-ized hydrogen is arbitrary and can prepare a coal-for-coke-making-ized hydrogen feed hopper according to the configuration of a fission reactor. For example, a coal-for-coke-making-ized hydrogen feed hopper may be prepared in the contraction section which may prepare a coal-for-coke-making-ized hydrogen feed hopper in the part from which the path of fission reactor 3a serves as max, and the path is reducing. Furthermore, as it \*\* to drawing 1, the coal-for-coke-making-ized hydrogen feed hopper 4 may be formed in the contraction section which the part from which the path of fission reactor 3a serves as max, and the path are reducing, respectively. The rate of flow of the gas in the location where coal-for-coke-making-ized hydrogen is introduced, the strength of a turbulent flow, etc. are controllable by the location of the coal-for-coke-making-ized hydrogen feed hopper 4.

[0036] As coal-for-coke-making-ized hydrogen, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially.

[0037] Two or more locations of the coal-for-coke-making-ized hydrogen feed hopper in a fission reactor may be prepared on the cross-section periphery of the flow direction of combustion gas, and the location which has two or more coal-for-coke-making-ized hydrogen feed hoppers on still such same periphery may be established in the flow direction of combustion gas multistage. In order to make generation reaction time of fullerene into homogeneity and for physical properties to obtain uniform fullerene, it is desirable to install as many coal-for-coke-making-ized hydrogen feed hoppers as possible

on the same periphery.

[0038] Moreover, although the form of the nozzle used for the coal-for-coke-making-ized hydrogen feed hopper 4 can be chosen suitably, when using the coal-for-coke-making-ized hydrogen of a liquid, in order to spray on homogeneity minutely more, it is desirable that the diameter of an initial drop of the coal-for-coke-making-ized hydrogen immediately after spraying from nozzles, such as 2 hydraulic nozzles which inject the supplied liquid with another liquid, considers as a small thing as much as possible. Although what is necessary is just to choose suitably, before the coal-for-coke-making-ized hydrogen sprayed on the 2nd reaction band 2 evaporates, as for the coal-for-coke-making-ized hydrogen supply approaches, such as a diameter of opening of the coal-for-coke-making-ized hydrogen feed hopper 4, a form, protrusion condition into a furnace, a supply include angle to a combustion gas style, and a gas-liquid ratio, the rate of flow, a flow rate, temperature, etc., it is desirable to spray on conditions which do not adhere to the furnace wall of the 2nd reaction band 2. By spraying such, the foreign matter in the soot-like matter obtained can be reduced.

[0039] The thing of arbitration can be used if it is the quality of the material which has thermal resistance, such as a metal and refractories, as internal insulation which constitutes the 1st reaction band 1 and the 2nd reaction band 2. Since the temperature of internal combustion gas becomes beyond metaled heat-resistant temperature when using a metal, it is necessary to cool from the outside by taking structures, such as rolling water cooled jacket structure and a water-cooled tube. As ingredients other than a metal, there are SiC, a diamond, nitriding aluminum, silicon nitride, ceramic system refractory material, etc., for example.

[0040] It is made into the structure which cools preferably 1000 degrees C or less of combustion gas styles containing the soot-like matter (the thing in the middle of a reaction is included) containing fullerene at 800 degrees C or less from the 2nd reaction band 2 after the downstream. Water etc. may be sprayed from a reaction halt fluid feed hopper, and, specifically, you may cool by passing the passage which cooled the exterior according to water-cooled structure etc. Especially, especially when the path of passage is small, even if it does not consider as water-cooled structure, it may fully be cooled by the natural heat dissipation to atmospheric air.

[0041] It dissociates with gas (not shown) and the fullerene and the soot-like matter which were cooled are recovered by the uptake bag filter prepared in the point of passage. The extraction approach of fullerene can use well-known general processes, such as making it adhere to such a bag filter or a passage wall etc.

[0042] As shown in drawing 2, the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 2nd of this invention The 1st reaction band 13 which the oxygen content gas and fuel gas which were supplied through the 1st burner 12 in the fission reactor 11 burn, and forms a hot combustion gas style, It is in the downstream of the 1st reaction band 13, and has the 2nd reaction band 16 which makes the coal-for-coke-making-ized hydrogen which has the delivery 15 of the 2nd burner 14 which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and was gasified and supplied react in a combustion gas style, and makes fullerene generate. Hereafter, these are explained to a detail. The fission reactor 11 is equipped with the cylindrical shape-like side-attachment-wall section 17 and the edge wall 19 which it connects with the end side of the side-attachment-wall section 17, and an outer diameter contracts gradually, and forms the exhaust port 18. The side-attachment-wall section 17 and the edge wall 19 consist of heat-resisting steel, such as stainless steel. Furthermore, the refractories which are not illustrated are lined by the inner skin by the side of the other end of the side-attachment-wall section 17. As refractories, the refractory brick of the quality of an alumina and the unshaped refractories of the quality of an alumina can be used, for example. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 18, and the other end side of an exhaust pipe is connected to the exhaust air pump. For this reason, while changing the inside of a fission reactor 11 into the reduced pressure condition of under atmospheric pressure, the combustion gas containing the soot-like matter generated in the fission reactor 11 can be discharged outside from the inside of a fission reactor 11.

[0043] The 1st burner 12 attached in base 17a by the side of the other end of the side-attachment-wall

section 17 has two or more oxygen content gas nozzles 21 linked to the oxygen content gas supply piping 20, and the fuel gas nozzle 23 linked to the fuel gas charging line 22, and mixture arrangement of each of these gas nozzles 21 and 23 is carried out at base 17a. Moreover, the oxygen content gas nozzle 21 and the fuel gas nozzle 23 are formed with heat-resisting steel, such as stainless steel. For this reason, after the oxygen content gas supplied from the oxygen content gas nozzle 21 and the fuel gas supplied from the fuel gas nozzle 23 are emitted, diffusive mixing of it will be carried out, it will be in the uniform mixed state, and burns in the 1st reaction band 13. And the formed hot combustion gas style flows into the 2nd reaction band 16 of the downstream. The 2nd burner 14 attached in the other end side of the side-attachment-wall section 17 consists of a minor diameter discharge tube 24 (for example, formed with heat-resisting steel, such as stainless steel) of a large number arranged by penetrating the 1st reaction band 13. Consequently, the delivery 15 established in the tip side of the minor diameter discharge tube 24 has a clearance in the upstream of the 2nd reaction band 16, and is arranged at it. Moreover, the end face side of each minor diameter discharge tube 24 is connected to the coal-for-coke-making-ized hydrogen charging line 25. For this reason, direct coal-for-coke-making-ized hydrogen can be supplied at homogeneity into the hot combustion gas style which flows from the 1st reaction band 13, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time.

[0044] Next, the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 2nd of this invention is explained to a detail. The fuel gas nozzle 23 to fuel gas is supplied for oxygen content gas from the oxygen content gas nozzle 21, a combustion gas style hot by burning these is formed, and it is made to circulate toward the lower stream of a river of a fission reactor 11. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a fission reactor 11, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature [ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of combustion gas.

[0045] What gasified coal system liquid fuel which gasified petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, such as a thing and creosote oil, as fuel gas can be used. Fuel gas, such as natural gas and petroleum gas, is desirable especially. Moreover, in order to gather the yield of fullerene, it is desirable to also dilute fuel gas using inert gas etc.

[0046] Then, the combustion gas style which fuel gas burns and forms under oxygen content gas is explained. While adjusting the amount of the fuel gas supplied from the fuel gas nozzle 23 on the conditions which fuel gas burns completely, and the amount of oxygen gas supplied from the oxygen content gas nozzle 21 and supplying the 1st reaction band 13, combustion of fuel gas starts with an ignition means to by\_which hold the inside of a fission reactor 11 and an exhaust-air pump does not illustrate it in the condition of 10 - 300torr more preferably under atmospheric pressure through the exhaust pipe which was connected to the exhaust port 18 and which is not illustrated. Here, fuel gas and oxygen content gas become independent respectively, and since it is emitted in the 1st reaction band 13 from the oxygen content gas nozzle 21 which separated distance and was distributed, and the fuel gas nozzle 23, they can make homogeneity the combustion condition in the 1st reaction band 13. Moreover, since the pressure in a fission reactor 11 has become under atmospheric pressure in addition to diluting with inert gas, such as argon gas, and falling, the oxygen gas concentration in oxygen content gas can change the combustion condition in the 1st reaction band 13 into the condition that it was similar with the elevated-temperature air combustion condition. Consequently, combustion of fuel gas advances to

homogeneity and can make temperature of the 1st reaction band 13 homogeneity and an elevated temperature (for example, 1000-1900 degrees C, preferably 1700-1900 degrees C).

[0047] Since the hot combustion gas formed in the 2nd reaction band 16 in the 1st reaction band 13 flows, the temperature of the upstream of the 2nd reaction band 16 becomes a 1000-1900-degree C elevated temperature. Distributed emission of the coal-for-coke-making-ized hydrogen is carried out into the combustion gas style of the upstream of the 2nd reaction band 16 from each delivery 15 of the minor diameter discharge tube 24 of a large number arranged by penetrating the 1st reaction band 13. Here, since the 1st reaction band 13 is penetrated and it is arranged, since the preheating is carried out while passing through the inside of the minor diameter discharge tube 24, the minor diameter discharge tube 24 pyrolyzes coal-for-coke-making-ized hydrogen, shortly after being emitted into a hot combustion gas style from a delivery 15. Consequently, the high pyrolysate of labile exists in combustion gas, and a fullerene precursor is formed when these coalesce. And it grows up, while a fullerene precursor moves with a combustion gas style, and it becomes fullerene. In addition, since the pyrolysis of coal-for-coke-making-ized hydrogen is endothermic reaction, heat energy is taken from combustion gas and the temperature of combustion gas falls. For this reason, oxygen content gas is mixed in coal-for-coke-making-ized hydrogen, a part of raw material carbon hydrogen is burned, and you may make it supply heat energy. however, a part of raw material carbon hydrogen -- since the ununiformity of the temperature in the 2nd reaction band 16 will arise and the generation effectiveness of fullerene will fall, if combustion takes place actively -- the oxygen density in combustion gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably.

[0048] As raw material carbon hydrogen, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of the raw material carbon hydrogen which mainly serves as a raw material uses an aromatic series system hydrocarbon especially.

[0049] As shown in drawing 3 , it is the description that premixing of oxygen content gas and the fuel gas is carried out, and the manufacturing installation 26 of the fullerene concerning the gestalt of operation of the 3rd of this invention is supplied to the 1st burner 27. Therefore, only the 1st burner 27 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. It is produced with the heat-resistant metal and the 1st burner 27 has the head 28 which the whole surface side has exposed to the 1st reaction band 13 of a fission reactor 11, and the accumulator 29 prepared in the lower part of a head 28. And each minor diameter discharge tube 24 of the 2nd burner 14 opened the predetermined clearance mutually, penetrated the accumulator 29 and the head 28 from the lower part of an accumulator 29, and has projected them in the fission reactor 11.

[0050] Here, the head 28 consists of porosity members of sintering metal. If the porosity member has structure equipped with many free passage holes which are open for free passage to a side on the other hand from the whole surface side, it considers as the mixed gas which carried out premixing of oxygen content gas and the fuel gas to the accumulator 29 prepared in the lower part of a head 28 and it supplies from the mixed-gas charging line 30 Mixed gas can be moved to the field exposed to the 1st reaction band 13 side from the field by the side of an accumulator 29 through the free passage hole in a head 28, and can be spouted in the 1st reaction band 13. Therefore, combustion gas hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the

delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-sized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing facility 26 of the fullerene concerning the gestalt of operation of the 3rd of this invention requires for the gestalt of the 2nd operation.

[0051] In the manufacturing installation 31 of the fullerene concerning the gestalt of operation of the 4th of this invention, since oxygen content gas and fuel gas are independently supplied to the 1st burner 32 for another piping, it is the description that the manufacturing installation 26 of the fullerene concerning the gestalt of the 3rd operation differs from the structure of the 1st burner 32. Therefore, only the 1st burner 32 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. That is, as shown in drawing 4, the 1st burner 32 is produced with a heat-resistant metal, and has two or more gas blenders 35 which have an exhaust nozzle in the head 33 which consists of a porosity member of sintering metallicity which has a free passage hole, the accumulator 34 prepared in the lower part of a head 33, and an accumulator 34. And each minor diameter discharge tube 24 of the 2nd burner 14 opened the predetermined clearance mutually, penetrated the accumulator 34 and the head 33 from the lower part of an accumulator 34, and has projected them in the fission reactor 11. Moreover, the aspirator-type mixer which attracts oxygen content gas and is mixed by the flow of fuel gas as a gas blender 35 can be used.

[0052] If oxygen content gas and fuel gas are independently supplied to each gas blender 35 by considering as such a configuration by the oxygen content gas supply piping 36 and the fuel gas charging line 37, respectively, oxygen content gas and fuel gas will flow in an accumulator 34 as mixed gas from the exhaust nozzle of a gas blender 35, being mixed. And the mixed gas which flowed in the accumulator 34 can be moved to the field exposed to the 1st reaction band 13 side from the field by the side of an accumulator 34 through the free passage hole in a head 33, and can be spouted in the 1st reaction band 13. Therefore, a combustion gas style hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-sized hydrogen supplied through the coal-for-coke-making-sized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-sized hydrogen can be pyrolyzed to homogeneity in a short time.

[0053] In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 26 of the fullerene which the manufacture approach of the fullerene which used the manufacturing facility 31 of the fullerene concerning the gestalt of operation of the 4th of this invention requires for the gestalt of the 3rd operation.

[0054] It be the description that the 1st burner 41 which have the header tubing 40 with which the jet nozzle 39 of the minor diameter of a large number which the mixed gas to which the manufacturing installation 38 of the fullerene which start the gestalt of operation of the 5th of this invention as show in drawing 5 be attached in base 17a by the side of the other end of the side attachment wall section 17, and premixing of oxygen content gas and the fuel gas be carried out spout set a clearance, and be form be supply. Therefore, only the 1st burner 41 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted.

[0055] The header tubing 40 had two or more circular canal 40a which prepared the clearance on this alignment, respectively and has been arranged to the axial center of a fission reactor 11, and has connected each circular canal 40a to mixed-gas charging line 30a. And through the clearance between each circular canal 40a, each minor diameter discharge tube 24 of the 2nd burner 14 penetrates the 1st reaction band 13, and is arranged. Therefore, if the mixed gas which carried out premixing of oxygen content gas and the fuel gas is supplied to each circular canal 40a through mixed-gas charging line 30a,

mixed gas will be spouted in the 1st reaction band 13 from each jet nozzle 39 of each circular canal 40a. For this reason, a combustion gas style hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing installation 38 of the fullerene concerning the gestalt of operation of the 5th of this invention requires for the gestalt of the 2nd operation.

[0056] As compared with the manufacturing installation 10 of the fullerene which the manufacturing installation 42 of the fullerene concerning the gestalt of operation of the 6th of this invention requires for the gestalt of operation of the 2nd of this invention, it is the description that the structures of the 1st burner 43 differ. Therefore, only the 1st burner 43 with which structures differ is explained, the same sign is given to the same component as the manufacturing installation 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. Namely, as shown in drawing 6, the 1st burner 43 attached in base 17a by the side of the other end of the side-attachment-wall section 17 is produced with a heat-resistant metal. It has the 2nd header tubing 47 with which the jet nozzle 46 of the minor diameter of a large number which the 1st header tubing 45 with which the jet nozzle 44 of the minor diameter of a large number which spout oxygen content gas set the clearance, and was formed, and the 1st header tubing 45 have a clearance, are arranged, and spout fuel gas set the clearance, and was formed. Furthermore, the oxygen content gas supply piping 20 and the fuel gas charging line 22 which supply independently oxygen content gas and said fuel gas, respectively are connected to the 1st header tubing 45 and the 2nd header tubing 47. Moreover, each minor diameter discharge tube 24 of the 2nd burner 14 penetrated base 17a through the clearance between the 1st header tubing 45 and the 2nd header tubing 47, and has projected it in the fission reactor 11.

[0057] Oxygen content gas can be supplied to the 1st header 45 through the oxygen content gas supply piping 20, and it can be made to blow off from the jet nozzle 44 in a fission reactor 11 by considering as such a configuration. Moreover, fuel gas can be supplied to the 2nd header 47 through the fuel gas charging line 22, and it can be made to blow off from the jet nozzle 46 in a fission reactor 11. After the oxygen content gas and fuel gas which blew off from each jet nozzles 44 and 46 are emitted, diffusive mixing of them will be carried out, they will be in the uniform mixed state, and burn in the 1st reaction band 13. And the formed hot combustion gas flows into the 2nd reaction band 16 of the downstream. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing installation 42 of the fullerene concerning the gestalt of operation of the 6th of this invention requires for the gestalt of the 2nd operation.

[0058] As mentioned above, although the gestalt of operation of this invention was explained, modification in the range which this invention is not limited to the gestalt of this operation, and does not change the summary of invention is possible, and also when it constitutes the manufacture approach of the fullerene of this invention, and its equipment combining the gestalt of each operation, or above mentioned a part or above mentioned all of a modification, it is the right range of this invention. For example, although constituted from two or more circular canal 40a arranged on this alignment to the axial center of a fission reactor 11 in the header tubing 40 with the gestalt of the 5th operation, a clearance may be prepared and two or more straight pipes may be arranged in in the shape of a grid, respectively. Moreover, although the clearance was prepared on this alignment to the axial center of a



fission reactor 11 and two or more 1st header tubing 45 and 2nd header tubing 47 have been arranged with the gestalt of the 6th operation, a clearance may be prepared and the 1st header tubing and the 2nd header tubing may be arranged in the shape of a grid, respectively. Furthermore, although the minor diameter discharge tube 24 of the 2nd burner 14 was produced with heat-resisting steel, such as stainless steel, and the porosity member was produced with the heat-resistant sintered metal with the gestalt of the 3rd and the 4th operation, it is also producible with a cermet and the ceramics.

[0059]

[Effect of the Invention] In the manufacture approach of fullerene according to claim 1 to 3 The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies the coal-for-coke-making-ized hydrogen gasified in the middle of this combustion gas style, and having the 2nd reaction band which makes coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. Since the pressure of the 2nd reaction band is made under into atmospheric pressure, the pyrolysis of coal-for-coke-making-ized hydrogen can advance to homogeneity, the generation effectiveness of fullerene can be raised, and fullerene can be manufactured cheaply and easily in large quantities.

[0060] On the other hand, in the manufacture approach of the fullerene by the above and the well-known combustion method, usual is the same and the fuel for a combustion reaction and the raw material for fullerene generation cannot select a fuel required for a hydrocarbon fuel combustion reaction to arbitration. On the other hand, since the fuel for a combustion reaction and the raw material for manufacture of fullerene can be selected separately according to this invention, when manufacturing fullerene especially on a scale of industry, the cheap original fuel of cost can be freely chosen according to the supply situation of a original fuel.

[0061] It sets to the manufacture approach of fullerene according to claim 2 especially. By being able to keep the conditions of the 2nd reaction band constant over all the cross sections in a furnace, and adjusting the conditions in this band on conditions from which the yield of fullerene serves as max, since the 2nd reaction band is in the downstream of the 1st reaction band Since the field which fullerene generates can be extended to max, compared with the usual combustion method, the yield of fullerene becomes high. On the other hand, although fullerene mainly generates in a flame in the conventional combustion method, generally, a flame has temperature distribution and it is known that fullerene will generate in the specific field of a flame.

[0062] In the manufacture approach of fullerene according to claim 3, since the temperature of the 2nd reaction band is 1000 degrees C or more, the pyrolysis of the supplied coal-for-coke-making-ized hydrogen can be carried out in a short time certainly, and fullerene can be manufactured in large quantities.

[0063] In the manufacturing installation of fullerene according to claim 4 to 13 The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, Since it has the 2nd reaction band which it is [ band ] in the downstream of the 1st reaction band, makes the coal-for-coke-making-ized hydrogen which has the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and was gasified and supplied react in a combustion gas style, and makes fullerene generate Both control of the combustion condition of a fuel and control of the pyrolysis of coal-for-coke-making-ized hydrogen become easy, and it becomes possible to manufacture fullerene in large quantities, cheaply, and easily.

[0064] Especially, in the manufacturing installation of fullerene according to claim 5, since the delivery of the 2nd burner has a clearance in the upstream of the 2nd reaction band, and a large number formation is carried out and it carries out distributed emission of the coal-for-coke-making-ized hydrogen into a combustion gas style, it can pyrolyze coal-for-coke-making-ized hydrogen to homogeneity in combustion gas in a short time, and becomes possible [ making high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen ].

[0065] In the manufacturing installation of fullerene according to claim 6 Since the 2nd burner consists



of a minor diameter discharge tube of a large number arranged by penetrating the 1st reaction band Distributed emission can be carried out uniformly, the coal-for-coke-making-ized hydrogen by which the preheating was carried out into the combustion gas style of the elevated temperature of the 2nd reaction band can be pyrolyzed, and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0066] In the manufacturing installation of fullerene according to claim 7, since the 1st burner emits oxygen content gas and fuel gas independently and it has two or more oxygen content gas nozzles and fuel gas nozzles by which mixture arrangement was carried out, you can carry out diffusive mixing of the oxygen content gas and fuel gas which were supplied, they can make it exist in the 1st reaction band by the uniform mixed state, and become possible [ carrying out the perfect combustion of the fuel gas easily in the 1st reaction band ]. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0067] In the manufacturing installation of fullerene according to claim 8, since it blows off where the head of the 1st burner consisted of a porosity member and oxygen content gas and fuel gas are mixed from a front face, oxygen content gas and fuel gas can be supplied to the 1st reaction band, where premixing is carried out, and it becomes possible to carry out the perfect combustion of the fuel gas easily in the 1st reaction band. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0068] In the manufacturing installation of fullerene according to claim 9, since mixing of oxygen content gas and fuel gas is performed within the 1st burner and oxygen content gas and fuel gas are independently supplied to the 1st burner for another piping, it is not necessary to establish the premixing means of oxygen content gas and fuel gas, and the configuration of the manufacturing installation of fullerene can be simplified.

[0069] In the manufacturing installation of fullerene according to claim 10, since the accumulator which premixing of oxygen content gas and the fuel gas was carried out, and was prepared in the lower part of a head is supplied, structure of the 1st burner can be simplified and the cost of the 1st burner can be reduced.

[0070] In the manufacturing installation of fullerene according to claim 11 the 1st burner Since the oxygen content gas and fuel gas by which have header tubing with which the jet nozzle of many minor diameters set the clearance, and was formed, and premixing was carried out to header tubing are supplied Where premixing is carried out, distributed emission of oxygen content gas and the fuel gas can be carried out in the 1st reaction band, and it becomes possible to carry out the perfect combustion of the fuel gas easily in the 1st reaction band. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0071] In the manufacturing installation of fullerene according to claim 12 The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout oxygen content gas set the clearance, and the 1st burner was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with the 1st header tubing, are arranged, and spout fuel gas set the clearance, and was formed. Since oxygen content gas and fuel gas are independently supplied to the 1st header tubing and the 2nd header tubing for another piping, respectively Diffusive mixing of the oxygen content gas and fuel gas by which distributed emission was carried out can be carried out, they can be in the uniform mixed state, can make it exist in the 1st reaction band, and become possible [ carrying out the perfect combustion of the fuel gas easily in the 1st reaction band ]. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0072] It can prevent that fill up the heat energy consumed when coal-for-coke-making-ized hydrogen pyrolyzed, and the temperature of combustion gas falls since oxygen content gas is mixed in the

manufacturing installation of fullerene according to claim 13 in the coal-for-coke-making-ized hydrogen supplied from the 2nd burner, and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

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[Translation done.]

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**DETAILED DESCRIPTION**

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[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the manufacture approach of fullerene, and its equipment.

[0002]

[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings so that it might be represented by C60 and C70 grade. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
- (3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below is mentioned as the manufacture approach of fullerene learned now.

(1) It is the approach of irradiating the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, and compounding by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot of laser is slight and it is unsuitable for extensive manufacture.

(2) It is the approach to which carry out energization heating and a graphite rod is made to sublime in the container under the reduced pressure filled with resistance heating method gaseous helium. Since the electric resistance loss in a circuit is large, it is unsuitable for extensive manufacture.

[0004] (3) It is the approach to which the carbon of a lifting and an anode plate is made to sublime arc

discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in number of arc discharge methods 10kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in a current works scale.

(4) Instead of using radio frequency heating method resistance heating and arc discharge, it is the approach of heating an eddy current to raw material graphite by RF induction, and evaporating a sink and raw material graphite.

(5) It is the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. It is observed as the mass-producing method for being a point usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc discharge method in the soot (fullerene etc.) which sub\*\* in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.

(6) It is the approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0005] Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene in large quantities cheaply by any approach until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, for example, the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the patent reference 1, and collecting condensates in it is indicated. This approach is an approach of manufacturing fullerene by burning a carbon inclusion in a flame, and the fuel for combustion and the raw material of fullerene are the same carbon inclusions substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black.

[0006] As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is industrially mentioned as the general manufacture approach. The carbon black manufacturing installation (fission reactor) of the shape for example, of a cylinder is used for this approach. To \*\*\*\*, to horizontal or a perpendicular direction, supply oxygen content gas and fuels, such as air, and they are burned in the 1st reaction band of the fission reactor concerned. It is made to move to the 2nd reaction band with the cross section which was installed in the lower stream of a river of furnace shaft orientations, and reduced the obtained combustion gas style. It is the approach of supplying coal-for-coke-making-sized hydrogen (stock oil), making it reacting into the gas stream concerned, making carbon black generating, quenching gas by spraying of cooling water etc. to a gas stream further in the 3rd reaction band on the lower stream of a river, and stopping a reaction.

[0007]

[Patent reference 1] \*\*\*\*\* No. 507879 [ six to ] official report [0008]

[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the above-mentioned usual carbon black, fullerene is hardly generated. In manufacture of fullerene, it has been a big technical problem how the rate of the fullerene contained in the soot-like matter obtained is raised. Generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [ these reduced pressure ] that diluent concentration will affect the yield of the above-mentioned fullerene.

[0009] In order to raise the yield of fullerene in the above and the patent reference 1, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating which heats a flame directly, microwave heating, discharge heating, and counterflow heating that heats a flame by heat exchange with elevated-temperature gas are mentioned.

[0010] By the above and the patent reference 1, pure oxygen is used as an oxidizer for a combustion reaction, and the argon is used as a diluent. This is considered to be effective in gathering the yield of fullerene. However, the amount of the oxygen needed for combustion also becomes extensive, and pure oxygen becomes a special oxygen supply facility is required and expensive [ the manufacturing cost of

fullerene ] as a result, when a bomb or supply equipment of dedication etc. tends to be required and it is going to manufacture fullerene on a scale of industry especially.

[0011] So, it has not result in utilization for the reasons of the rate which the volume increases at the time of the actuation under that combustion temperature becomes low since there are many rates of that a flame is not stabilize compared with pure oxygen since the oxygen density is low although it can guess easily use air as an oxidizer of combustion in order to reduce a manufacturing cost in a combustion method, or nitrogen, especially reduced pressure, and passes a nozzle become quick. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the technique of manufacturing fullerene cheaply and easily in large quantities is desired.

[0012] This invention is made in view of a situation which was mentioned above, and it aims at offering the manufacture approach of the fullerene which manufacture fullerene cheaply and easily in large quantities, and its equipment.

[0013]

[Means for Solving the Problem] The first reaction band in which this invention persons supply oxygen content gas and a fuel, and burn them in a fission reactor as a result of examining various the optimal combustion methods and manufacturing installations which can manufacture fullerene in large quantities and cheaply, and a hot combustion gas style is made to form, The manufacturing installation of the fullerene which have the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and have the 2nd reaction band which makes coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. Knowledge that fullerene is stably generable in large quantities by maintaining the pressure of the 2nd reaction band under at atmospheric pressure was acquired.

[0014] Namely, the manufacture approach of the fullerene concerning the 1st invention in alignment with said purpose The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the middle of this combustion gas style, and having the 2nd reaction band which makes this coal-for-coke-making-ized hydrogen react and makes fullerene generate is used, and the pressure of said 2nd reaction band is made under into atmospheric pressure. Since a fuel and oxygen content gas are supplied and are burned in the 1st reaction band, perfect combustion can be attained easily, for example and a hot combustion gas style can be formed. And by supplying coal-for-coke-making-ized hydrogen into the acquired hot gas stream, the pyrolysis of the coal-for-coke-making-ized hydrogen can be carried out easily, and the generation effectiveness of fullerene can be raised. Moreover, by making the pressure in the 2nd reaction band under into atmospheric pressure, and rarefying the mixed state of coal-for-coke-making-ized hydrogen and combustion gas, the pyrolysis of coal-for-coke-making-ized hydrogen can advance to homogeneity and the generation effectiveness of fullerene can be raised.

[0015] In the manufacture approach of the fullerene concerning the 1st invention, it is desirable that said 2nd reaction band is in the downstream of said 1st reaction band. By establishing the 2nd reaction band in the downstream of the 1st reaction band, the hot combustion gas formed in the 1st reaction band can be immediately introduced into the 2nd reaction band. Consequently, temperature of the 2nd reaction band can be made into an elevated temperature. In the manufacture approach of the fullerene concerning the 1st invention, it is desirable that the temperature of said 2nd reaction band is 1000 degrees C or more. By making temperature of the 2nd reaction band into 1000 degrees C or more, the pyrolysis of the supplied coal-for-coke-making-ized hydrogen can be carried out in a short time certainly.

[0016] The manufacturing installation of the fullerene concerning the 2nd invention in alignment with said purpose The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, It is in the downstream of this 1st reaction band, and has the 2nd reaction band which makes said coal-for-coke-making-ized hydrogen which has the delivery of the 2nd burner which supplies coal-for-coke-making-

ized hydrogen in the style of [ said ] combustion gas, and was gasified and supplied react in said combustion gas style, and makes fullerene generate. Since combustion of a fuel is performed in the 1st reaction band, control of a combustion condition becomes easy and hot combustion gas can be formed easily. Control of the pyrolysis of coal-for-coke-making-ized hydrogen becomes easy by introducing the obtained hot combustion gas style into the 2nd reaction band, and adjusting gas stream conditions, such as temperature of a hot combustion gas style, the rate of flow, and a flow rate, and the conditions of supply of coal-for-coke-making-ized hydrogen, since the pyrolysis of the coal-for-coke-making-ized hydrogen is supplied and carried out into this hot gas stream.

[0017] As for the delivery of said 2nd burner, in the manufacturing installation of the fullerene concerning the 2nd invention, it is desirable to have a clearance in the upstream of said 2nd reaction band, and for a large number formation to be carried out and to carry out distributed emission of said coal-for-coke-making-ized hydrogen into said combustion gas style. By forming in the upstream of the 2nd reaction band the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen, direct coal-for-coke-making-ized hydrogen can be supplied into the hot combustion gas style which flows from the 1st reaction band, and the pyrolysis of the coal-for-coke-making-ized hydrogen can be carried out easily. Moreover, since distributed emission of the coal-for-coke-making-ized hydrogen is carried out into combustion gas from many deliveries, coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in combustion gas in a short time. As for said 2nd burner, in the manufacturing installation of the fullerene concerning the 2nd invention, it is desirable to consist of a minor diameter discharge tube of a large number arranged by penetrating said 1st reaction band. Since coal-for-coke-making-ized hydrogen is supplied with many minor diameter discharge tubes, distributed emission of the coal-for-coke-making-ized hydrogen can be uniformly carried out into the combustion gas style of the elevated temperature of the 2nd reaction band. Moreover, since a minor diameter discharge tube penetrates the 1st reaction band and is arranged, coal-for-coke-making-ized hydrogen is gradually heated by hot combustion gas, passing through the inside of a minor diameter discharge tube, and can promote the pyrolysis in the inside of the combustion gas style of the elevated temperature of the 2nd reaction band.

[0018] In the manufacturing installation of the fullerene concerning the 2nd invention, mixture arrangement of two or more oxygen content gas nozzles and fuel gas nozzles to which said 1st burner emits independently said oxygen content gas and said fuel gas, respectively may be carried out. Diffusive mixing of the oxygen content gas and fuel gas which were supplied can be carried out, they can be in the uniform mixed state, and can be made to exist in the 1st reaction band by considering as such a configuration. Moreover, in the manufacturing installation of the fullerene concerning the 2nd invention, the head of said 1st burner consists of a porosity member, and can be considered as the configuration which blows off from a front face where said oxygen content gas and said fuel gas are mixed. By considering as such a configuration, oxygen content gas and fuel gas can be supplied to the 1st reaction band, where premixing is carried out.

[0019] In the manufacturing installation of the fullerene concerning the 2nd invention, mixing of said oxygen content gas and said fuel gas is performed within said 1st burner, and it can consider as the configuration to which said oxygen content gas and said fuel gas are independently supplied for another piping at said 1st burner. Since mixing of oxygen content gas and fuel gas is performed within the 1st burner, it is not necessary to establish separately the premixing means of oxygen content gas and fuel gas, and the configuration of the manufacturing installation of fullerene becomes easy. In the manufacturing installation of the fullerene concerning the 2nd invention, said oxygen content gas and said fuel gas can be considered as the configuration supplied to the accumulator which premixing was carried out and was prepared in the lower part of said head. Since premixing of oxygen content gas and the fuel gas is carried out and they are supplied to the accumulator of the lower part of a head, structure of the 1st burner can be simplified.

[0020] In the manufacturing installation of the fullerene concerning the 2nd invention, said 1st burner has header tubing with which the jet nozzle of many minor diameters set the clearance, and was formed, and can consider it as the configuration to which said oxygen content gas with which premixing was

carried out to this header tubing, and said fuel gas are supplied. By considering as such a configuration, where premixing is carried out, distributed emission of oxygen content gas and the fuel gas can be carried out in the 1st reaction band. In the manufacturing installation of the fullerene concerning the 2nd invention said 1st burner The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout said oxygen content gas set the clearance, and was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with said 1st header tubing, are arranged, and spout said fuel gas set the clearance, and was formed. It can consider as the configuration to which said oxygen content gas and said fuel gas are independently supplied for another piping, respectively at said 1st header tubing and said 2nd header tubing. Diffusive mixing of the oxygen content gas and fuel gas by which distributed emission was carried out can be carried out, they can be in the uniform mixed state, and can be made to exist in the 1st reaction band by considering as such a configuration.

[0021] In the manufacturing installation of the fullerene concerning the 2nd invention, oxygen content gas is mixable in the coal-for-coke-making-ized hydrogen supplied from said 2nd burner. In the pyrolysis of coal-for-coke-making-ized hydrogen, the temperature of combustion gas falls by the pyrolysis of coal-for-coke-making-ized hydrogen for endothermic reaction. For this reason, it can prevent that fill up the heat energy consumed when a part of coal-for-coke-making-ized hydrogen was burned in the 2nd reaction band, heat energy was generated and coal-for-coke-making-ized hydrogen pyrolyzed by mixing oxygen content gas in coal-for-coke-making-ized hydrogen, and the temperature of combustion gas falls.

[0022]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented. The explanatory view of the fullerene manufacturing installation which applied the manufacture approach of the fullerene which drawing 1 (A) and (B) require for the gestalt of operation of the 1st of this invention, respectively here, The explanatory view of the manufacturing installation of the fullerene which a plane section Fig., drawing 2 (A), and (B) require for the gestalt of operation of the 2nd of this invention, respectively, The explanatory view of the manufacturing installation of the fullerene which a plane section Fig., drawing 3 (A), and (B) require for the gestalt of operation of the 3rd of this invention, respectively, A plane section Fig., the partial explanatory view of the manufacturing installation of the fullerene which drawing 4 requires for the gestalt of operation of the 4th of this invention, The explanatory view of the manufacturing installation of the fullerene which drawing 5 (A) and (B) require for the gestalt of operation of the 5th of this invention, respectively, a plane section Fig., drawing 6 (A), and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 6th of this invention, and a plane section Fig., respectively.

[0023] The manufacture approach of the fullerene concerning the gestalt of operation of the 1st of this invention is explained using drawing 1 . The manufacture approach of the fullerene concerning the gestalt of the 1st operation is related with the approach of introducing coal-for-coke-making-ized hydrogen into the manufacturing installation 3 of the fullerene constituted by forming the 1st reaction band 1 and the 2nd reaction band 2 in fission reactor 3a, and manufacturing fullerene by burning.

[0024] The manufacturing installation 3 of fullerene has the 2nd reaction band 2 which coal-for-coke-making-ized hydrogen is supplied [ band ], makes it react the 1st reaction band 1 in which a combustion gas style is made to form, and in the style of [ which were formed there ] combustion gas, and makes fullerene generate. The 2nd reaction band 2 may be in the downstream of the direction of a combustion gas style (it may be hereafter called "shaft orientations") which may be the almost same field (an outside or inside) as the 1st reaction band 1, and was formed in the 1st reaction band 1.

[0025] Drawing 1 shows the case where the 2nd reaction band 2 is located on the lower stream of a river of the 1st reaction band 1.

Generally in the [1st reaction band] 1st reaction band 1, a combustion gas style hot by supplying a fuel and oxygen content gas and burning them, respectively is generated toward the lower stream of a river of the 2nd reaction band 2, i.e., fission reactor 3a, from a fuel feed hopper and oxygen content gas

supply opening.

[0026] Even if supply of a fuel and oxygen content gas is the so-called premixing method mixed before entering in fission reactor 3a, it may be the so-called diffusive-mixing method supplied to fission reactor 3a from the nozzle which became independent, respectively. In drawing 1, in the case of a diffusive-mixing method, a fuel is supplied from the central fuel feed hopper 7, and it supplies oxygen content gas from the oxygen content gas supply openings 5 and 6 of the perimeter. Moreover, a premixing method and a diffusive-mixing method may be combined, for example, in drawing 1, from the oxygen content gas supply opening 5, what mixed oxygen content gas with the fuel beforehand may be supplied, and the fuel from the fuel feed hopper 7 may be independently supplied for oxygen content gas from the oxygen content gas supply opening 6, respectively.

[0027] It may be the purpose that this 1st reaction band 1 generates hot combustion gas, and that combustion method may be what kind of well-known combustion methods, such as premixed combustion, diffusive burning, laminar-flow combustion, turbulent flow combustion, and elevated-temperature air combustion. Moreover, although combustion in the 1st reaction band 1 may be perfect combustion or you may be incomplete combustion as long as the temperature which becomes generable [ fullerene ] in the 2nd reaction band 2 is acquired, it is desirable that it is perfect combustion with the large calorific value to fuel used. When the 1st reaction band 1 is incomplete combustion with the so-called superfluous fuel, the soot-like matter which contains fullerene even in the 1st reaction band 1 may generate.

[0028] However, the combustion by the lean mixture whose oxygen required for combustion is more than the amount of stoichiometry oxygen of the combustion in this 1st reaction band 1 is preferably better. As oxygen content gas, the gas which mixed non-flammable gas, such as argon gas and nitrogen gas, at a rate of arbitration can be used for air, oxygen gas, or these. NOX especially in elevated-temperature combustion Pure oxygen may be used in order to suppress generating. In order to gather the yield of fullerene, it is desirable to dilute using rare gas etc. in a combustion process. Rare gas may be supplied from the exclusive nozzle for supply, and may be beforehand mixed in a fuel, coal-for-coke-making-ized hydrogen, and oxygen content gas.

[0029] As a fuel, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, a fuel oil, benzene, and toluene, and creosote oil, can be used. Especially, as a fuel used with the gestalt of this operation, fuel gas is desirable. Moreover, although what is necessary is for fullerene to obtain just to adjust suitably the mean temperature in the 1st reaction band 1 at the time of fullerene manufacture, it is preferably made into 1600 degrees C or more still more preferably 1300 degrees C or more. This is because the productivity of fullerene goes up, so that the temperature of combustion gas is an elevated temperature. Even if an upper limit is too high not much, the productivity of fullerene may fall. Moreover, what is necessary is just to determine after taking into consideration the heat-resistant problem by the quality of the material of a fission reactor.

[0030] If opening of the arrangement of the fuel feed hopper 7 and the oxygen content gas supply openings 5 and 6 is carried out to fission reactor 3a, it is arbitrary. In drawing 1, opening of the fuel feed hopper 7 and the oxygen content gas supply openings 5 and 6 is carried out to the same fission reactor 3a side. The configuration of each feed hoppers 5, 6, and 7 which are carrying out opening into fission reactor 3a may be arbitrary, and may be the indeterminate form of the shape of a polygon, such as an approximate circle form, an ellipse form, and the shape of a trigonum and a rectangular head, a gourd mold, etc.

[0031] As for fission reactor 3a internal pressure, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

Coal-for-coke-making-ized hydrogen is supplied from the coal-for-coke-making-ized hydrogen feed hopper 4 in the style of [ which was formed in the 1st reaction band 1 ] combustion gas, and fullerene is made to generate in the [2nd reaction band] 2nd reaction band 2 by carrying out partial combustion of a part of this coal-for-coke-making-ized hydrogen. In order to carry out partial combustion, it is good also considering the combustion in the 1st reaction band 1 as hyperoxia so that oxygen may remain.



Moreover, a nozzle may be arranged to the 2nd reaction field 2, and oxygen content gas may be supplied to it from an oxygen content gas supply nozzle.

[0032] Under the present circumstances, as for the above-mentioned coal-for-coke-making-ized hydrogen supplied into combustion gas, or oxygen content gas, it is desirable to be supplied in fission reactor 3a as much as possible at homogeneity. For this reason, it is desirable to be equally arranged so well that many by the number of the coal-for-coke-making-ized hydrogen feed hopper 4 installed in the 2nd reaction band 2 and an oxygen content gas supply nozzle in fission reactor 3a.

[0033] What is necessary is just to choose the die length of the 2nd reaction band suitably according to the magnitude of fission reactor 3a, the class of fullerene to manufacture, etc. The location and configuration of the 2nd reaction band may be arbitrary, and may be the inside of the 1st reaction band, or may be an outside, and as shown in drawing 1, they may be in the downstream of the 1st reaction band 1. It is more desirable for the cross-section configuration of the 2nd reaction band not to change, although the configuration of the 2nd reaction band is also arbitrary. The reason is that it will have effect which is not desirable on the fullerene to generate if influenced by the flow by the cross-section configuration of the 2nd reaction band changing in the process which fullerene generates of turbulence.

[0034] Although what is necessary is just to choose the mean temperature of the 2nd reaction band 2 suitably by the fullerene to manufacture, in order that coal-for-coke-making-ized hydrogen may evaporate and react to homogeneity, it is desirable that it is an elevated-temperature ambient atmosphere enough. It is desirable that it is specifically 1000 degrees C or more, and it is especially desirable that it is 1700-1900 degrees C especially. Moreover, in the 2nd reaction band 2, it is desirable to control the oxygen density in combustion gas as much as possible. It is because there is a thing of coal-for-coke-making-ized hydrogen, the generation reaction band 2, i.e., 2nd reaction band, of fullerene, which combustion takes place actively in part, therefore the ununiformity of the temperature in the 2nd reaction band 2 produces when oxygen exists so much in combustion gas. the oxygen density in combustion gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably.

[0035] In the gestalt of this operation, the location which supplies coal-for-coke-making-ized hydrogen is arbitrary and can prepare a coal-for-coke-making-ized hydrogen feed hopper according to the configuration of a fission reactor. For example, a coal-for-coke-making-ized hydrogen feed hopper may be prepared in the contraction section which may prepare a coal-for-coke-making-ized hydrogen feed hopper in the part from which the path of fission reactor 3a serves as max, and the path is reducing. Furthermore, as it \*\* to drawing 1, the coal-for-coke-making-ized hydrogen feed hopper 4 may be formed in the contraction section which the part from which the path of fission reactor 3a serves as max, and the path are reducing, respectively. The rate of flow of the gas in the location where coal-for-coke-making-ized hydrogen is introduced, the strength of a turbulent flow, etc. are controllable by the location of the coal-for-coke-making-ized hydrogen feed hopper 4.

[0036] As coal-for-coke-making-ized hydrogen, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially.

[0037] Two or more locations of the coal-for-coke-making-ized hydrogen feed hopper in a fission reactor may be prepared on the cross-section periphery of the flow direction of combustion gas, and the location which has two or more coal-for-coke-making-ized hydrogen feed hoppers on still such same periphery may be established in the flow direction of combustion gas multistage. In order to make generation reaction time of fullerene into homogeneity and for physical properties to obtain uniform fullerene, it is desirable to install as many coal-for-coke-making-ized hydrogen feed hoppers as possible

on the same periphery.

[0038] Moreover, although the form of the nozzle used for the coal-for-coke-making-ized hydrogen feed hopper 4 can be chosen suitably, when using the coal-for-coke-making-ized hydrogen of a liquid, in order to spray on homogeneity minutely more, it is desirable that the diameter of an initial drop of the coal-for-coke-making-ized hydrogen immediately after spraying from nozzles, such as 2 hydraulic nozzles which inject the supplied liquid with another liquid, considers as a small thing as much as possible. Although what is necessary is just to choose suitably, before the coal-for-coke-making-ized hydrogen sprayed on the 2nd reaction band 2 evaporates, as for the coal-for-coke-making-ized hydrogen supply approaches, such as a diameter of opening of the coal-for-coke-making-ized hydrogen feed hopper 4, a form, protrusion condition into a furnace, a supply include angle to a combustion gas style, and a gas-liquid ratio, the rate of flow, a flow rate, temperature, etc., it is desirable to spray on conditions which do not adhere to the furnace wall of the 2nd reaction band 2. By spraying such, the foreign matter in the soot-like matter obtained can be reduced.

[0039] The thing of arbitration can be used if it is the quality of the material which has thermal resistance, such as a metal and refractories, as internal insulation which constitutes the 1st reaction band 1 and the 2nd reaction band 2. Since the temperature of internal combustion gas becomes beyond metaled heat-resistant temperature when using a metal, it is necessary to cool from the outside by taking structures, such as rolling water cooled jacket structure and a water-cooled tube. As ingredients other than a metal, there are SiC, a diamond, nitriding aluminum, silicon nitride, ceramic system refractory material, etc., for example.

[0040] It is made into the structure which cools preferably 1000 degrees C or less of combustion gas styles containing the soot-like matter (the thing in the middle of a reaction is included) containing fullerene at 800 degrees C or less from the 2nd reaction band 2 after the downstream. Water etc. may be sprayed from a reaction halt fluid feed hopper, and, specifically, you may cool by passing the passage which cooled the exterior according to water-cooled structure etc. Especially, especially when the path of passage is small, even if it does not consider as water-cooled structure, it may fully be cooled by the natural heat dissipation to atmospheric air.

[0041] It dissociates with gas (not shown) and the fullerene and the soot-like matter which were cooled are recovered by the uptake bag filter prepared in the point of passage. The extraction approach of fullerene can use well-known general processes, such as making it adhere to such a bag filter or a passage wall etc.

[0042] As shown in drawing 2, the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 2nd of this invention The 1st reaction band 13 which the oxygen content gas and fuel gas which were supplied through the 1st burner 12 in the fission reactor 11 burn, and forms a hot combustion gas style, It is in the downstream of the 1st reaction band 13, and has the 2nd reaction band 16 which makes the coal-for-coke-making-ized hydrogen which has the delivery 15 of the 2nd burner 14 which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and was gasified and supplied react in a combustion gas style, and makes fullerene generate. Hereafter, these are explained to a detail. The fission reactor 11 is equipped with the cylindrical shape-like side-attachment-wall section 17 and the edge wall 19 which it connects with the end side of the side-attachment-wall section 17, and an outer diameter contracts gradually, and forms the exhaust port 18. The side-attachment-wall section 17 and the edge wall 19 consist of heat-resisting steel, such as stainless steel. Furthermore, the refractories which are not illustrated are lined by the inner skin by the side of the other end of the side-attachment-wall section 17. As refractories, the refractory brick of the quality of an alumina and the unshaped refractories of the quality of an alumina can be used, for example. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 18, and the other end side of an exhaust pipe is connected to the exhaust air pump. For this reason, while changing the inside of a fission reactor 11 into the reduced pressure condition of under atmospheric pressure, the combustion gas containing the soot-like matter generated in the fission reactor 11 can be discharged outside from the inside of a fission reactor 11.

[0043] The 1st burner 12 attached in base 17a by the side of the other end of the side-attachment-wall

section 17 has two or more oxygen content gas nozzles 21 linked to the oxygen content gas supply piping 20, and the fuel gas nozzle 23 linked to the fuel gas charging line 22, and mixture arrangement of each of these gas nozzles 21 and 23 is carried out at base 17a. Moreover, the oxygen content gas nozzle 21 and the fuel gas nozzle 23 are formed with heat-resisting steel, such as stainless steel. For this reason, after the oxygen content gas supplied from the oxygen content gas nozzle 21 and the fuel gas supplied from the fuel gas nozzle 23 are emitted, diffusive mixing of it will be carried out, it will be in the uniform mixed state, and burns in the 1st reaction band 13. And the formed hot combustion gas style flows into the 2nd reaction band 16 of the downstream. The 2nd burner 14 attached in the other end side of the side-attachment-wall section 17 consists of a minor diameter discharge tube 24 (for example, formed with heat-resisting steel, such as stainless steel) of a large number arranged by penetrating the 1st reaction band 13. Consequently, the delivery 15 established in the tip side of the minor diameter discharge tube 24 has a clearance in the upstream of the 2nd reaction band 16, and is arranged at it. Moreover, the end face side of each minor diameter discharge tube 24 is connected to the coal-for-coke-making-ized hydrogen charging line 25. For this reason, direct coal-for-coke-making-ized hydrogen can be supplied at homogeneity into the hot combustion gas style which flows from the 1st reaction band 13, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time.

[0044] Next, the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 2nd of this invention is explained to a detail. The fuel gas nozzle 23 to fuel gas is supplied for oxygen content gas from the oxygen content gas nozzle 21, a combustion gas style hot by burning these is formed, and it is made to circulate toward the lower stream of a river of a fission reactor 11. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a fission reactor 11, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature [ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of combustion gas.

[0045] What gasified coal system liquid fuel which gasified petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, such as a thing and creosote oil, as fuel gas can be used. Fuel gas, such as natural gas and petroleum gas, is desirable especially. Moreover, in order to gather the yield of fullerene, it is desirable to also dilute fuel gas using inert gas etc.

[0046] Then, the combustion gas style which fuel gas burns and forms under oxygen content gas is explained. While adjusting the amount of the fuel gas supplied from the fuel gas nozzle 23 on the conditions which fuel gas burns completely, and the amount of oxygen gas supplied from the oxygen content gas nozzle 21 and supplying the 1st reaction band 13, combustion of fuel gas starts with an ignition means to by\_which hold the inside of a fission reactor 11 and an exhaust-air pump does not illustrate it in the condition of 10 - 300torr more preferably under atmospheric pressure through the exhaust pipe which was connected to the exhaust port 18 and which is not illustrated. Here, fuel gas and oxygen content gas become independent respectively, and since it is emitted in the 1st reaction band 13 from the oxygen content gas nozzle 21 which separated distance and was distributed, and the fuel gas nozzle 23, they can make homogeneity the combustion condition in the 1st reaction band 13. Moreover, since the pressure in a fission reactor 11 has become under atmospheric pressure in addition to diluting with inert gas, such as argon gas, and falling, the oxygen gas concentration in oxygen content gas can change the combustion condition in the 1st reaction band 13 into the condition that it was similar with the elevated-temperature air combustion condition. Consequently, combustion of fuel gas advances to

homogeneity and can make temperature of the 1st reaction band 13 homogeneity and an elevated temperature (for example, 1000-1900 degrees C, preferably 1700-1900 degrees C).

[0047] Since the hot combustion gas formed in the 2nd reaction band 16 in the 1st reaction band 13 flows, the temperature of the upstream of the 2nd reaction band 16 becomes a 1000-1900-degree C elevated temperature. Distributed emission of the coal-for-coke-making-ized hydrogen is carried out into the combustion gas style of the upstream of the 2nd reaction band 16 from each delivery 15 of the minor diameter discharge tube 24 of a large number arranged by penetrating the 1st reaction band 13. Here, since the 1st reaction band 13 is penetrated and it is arranged, since the preheating is carried out while passing through the inside of the minor diameter discharge tube 24, the minor diameter discharge tube 24 pyrolyzes coal-for-coke-making-ized hydrogen, shortly after being emitted into a hot combustion gas style from a delivery 15. Consequently, the high pyrolysate of labile exists in combustion gas, and a fullerene precursor is formed when these coalesce. And it grows up, while a fullerene precursor moves with a combustion gas style, and it becomes fullerene. In addition, since the pyrolysis of coal-for-coke-making-ized hydrogen is endothermic reaction, heat energy is taken from combustion gas and the temperature of combustion gas falls. For this reason, oxygen content gas is mixed in coal-for-coke-making-ized hydrogen, a part of raw material carbon hydrogen is burned, and you may make it supply heat energy. however, a part of raw material carbon hydrogen -- since the ununiformity of the temperature in the 2nd reaction band 16 will arise and the generation effectiveness of fullerene will fall, if combustion takes place actively -- the oxygen density in combustion gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably.

[0048] As raw material carbon hydrogen, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of the raw material carbon hydrogen which mainly serves as a raw material uses an aromatic series system hydrocarbon especially.

[0049] As shown in drawing 3 , it is the description that premixing of oxygen content gas and the fuel gas is carried out, and the manufacturing installation 26 of the fullerene concerning the gestalt of operation of the 3rd of this invention is supplied to the 1st burner 27. Therefore, only the 1st burner 27 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. It is produced with the heat-resistant metal and the 1st burner 27 has the head 28 which the whole surface side has exposed to the 1st reaction band 13 of a fission reactor 11, and the accumulator 29 prepared in the lower part of a head 28. And each minor diameter discharge tube 24 of the 2nd burner 14 opened the predetermined clearance mutually, penetrated the accumulator 29 and the head 28 from the lower part of an accumulator 29, and has projected them in the fission reactor 11.

[0050] Here, the head 28 consists of porosity members of sintering metal. If the porosity member has structure equipped with many free passage holes which are open for free passage to a side on the other hand from the whole surface side, it considers as the mixed gas which carried out premixing of oxygen content gas and the fuel gas to the accumulator 29 prepared in the lower part of a head 28 and it supplies from the mixed-gas charging line 30 Mixed gas can be moved to the field exposed to the 1st reaction band 13 side from the field by the side of an accumulator 29 through the free passage hole in a head 28, and can be spouted in the 1st reaction band 13. Therefore, combustion gas hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the

delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing facility 26 of the fullerene concerning the gestalt of operation of the 3rd of this invention requires for the gestalt of the 2nd operation.

[0051] In the manufacturing installation 31 of the fullerene concerning the gestalt of operation of the 4th of this invention, since oxygen content gas and fuel gas are independently supplied to the 1st burner 32 for another piping, it is the description that the manufacturing installation 26 of the fullerene concerning the gestalt of the 3rd operation differs from the structure of the 1st burner 32. Therefore, only the 1st burner 32 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. That is, as shown in drawing 4, the 1st burner 32 is produced with a heat-resistant metal, and has two or more gas blenders 35 which have an exhaust nozzle in the head 33 which consists of a porosity member of sintering metallicity which has a free passage hole, the accumulator 34 prepared in the lower part of a head 33, and an accumulator 34. And each minor diameter discharge tube 24 of the 2nd burner 14 opened the predetermined clearance mutually, penetrated the accumulator 34 and the head 33 from the lower part of an accumulator 34, and has projected them in the fission reactor 11. Moreover, the aspirator-type mixer which attracts oxygen content gas and is mixed by the flow of fuel gas as a gas blender 35 can be used.

[0052] If oxygen content gas and fuel gas are independently supplied to each gas blender 35 by considering as such a configuration by the oxygen content gas supply piping 36 and the fuel gas charging line 37, respectively, oxygen content gas and fuel gas will flow in an accumulator 34 as mixed gas from the exhaust nozzle of a gas blender 35, being mixed. And the mixed gas which flowed in the accumulator 34 can be moved to the field exposed to the 1st reaction band 13 side from the field by the side of an accumulator 34 through the free passage hole in a head 33, and can be spouted in the 1st reaction band 13. Therefore, a combustion gas style hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time.

[0053] In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 26 of the fullerene which the manufacture approach of the fullerene which used the manufacturing facility 31 of the fullerene concerning the gestalt of operation of the 4th of this invention requires for the gestalt of the 3rd operation.

[0054] It be the description that the 1st burner 41 which have the header tubing 40 with which the jet nozzle 39 of the minor diameter of a large number which the mixed gas to which the manufacturing installation 38 of the fullerene which start the gestalt of operation of the 5th of this invention as show in drawing 5 be attached in base 17a by the side of the other end of the side attachment wall section 17, and premixing of oxygen content gas and the fuel gas be carried out spout set a clearance, and be form be supply. Therefore, only the 1st burner 41 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted.

[0055] The header tubing 40 had two or more circular canal 40a which prepared the clearance on this alignment, respectively and has been arranged to the axial center of a fission reactor 11, and has connected each circular canal 40a to mixed-gas charging line 30a. And through the clearance between each circular canal 40a, each minor diameter discharge tube 24 of the 2nd burner 14 penetrates the 1st reaction band 13, and is arranged. Therefore, if the mixed gas which carried out premixing of oxygen content gas and the fuel gas is supplied to each circular canal 40a through mixed-gas charging line 30a,

mixed gas will be spouted in the 1st reaction band 13 from each jet nozzle 39 of each circular canal 40a. For this reason, a combustion gas style hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing installation 38 of the fullerene concerning the gestalt of operation of the 5th of this invention requires for the gestalt of the 2nd operation.

[0056] As compared with the manufacturing installation 10 of the fullerene which the manufacturing installation 42 of the fullerene concerning the gestalt of operation of the 6th of this invention requires for the gestalt of operation of the 2nd of this invention, it is the description that the structures of the 1st burner 43 differ. Therefore, only the 1st burner 43 with which structures differ is explained, the same sign is given to the same component as the manufacturing installation 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. Namely, as shown in drawing 6, the 1st burner 43 attached in base 17a by the side of the other end of the side-attachment-wall section 17 is produced with a heat-resistant metal. It has the 2nd header tubing 47 with which the jet nozzle 46 of the minor diameter of a large number which the 1st header tubing 45 with which the jet nozzle 44 of the minor diameter of a large number which spout oxygen content gas set the clearance, and was formed, and the 1st header tubing 45 have a clearance, are arranged, and spout fuel gas set the clearance, and was formed. Furthermore, the oxygen content gas supply piping 20 and the fuel gas charging line 22 which supply independently oxygen content gas and said fuel gas, respectively are connected to the 1st header tubing 45 and the 2nd header tubing 47. Moreover, each minor diameter discharge tube 24 of the 2nd burner 14 penetrated base 17a through the clearance between the 1st header tubing 45 and the 2nd header tubing 47, and has projected it in the fission reactor 11.

[0057] Oxygen content gas can be supplied to the 1st header 45 through the oxygen content gas supply piping 20, and it can be made to blow off from the jet nozzle 44 in a fission reactor 11 by considering as such a configuration. Moreover, fuel gas can be supplied to the 2nd header 47 through the fuel gas charging line 22, and it can be made to blow off from the jet nozzle 46 in a fission reactor 11. After the oxygen content gas and fuel gas which blew off from each jet nozzles 44 and 46 are emitted, diffusive mixing of them will be carried out, they will be in the uniform mixed state, and burn in the 1st reaction band 13. And the formed hot combustion gas flows into the 2nd reaction band 16 of the downstream. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing installation 42 of the fullerene concerning the gestalt of operation of the 6th of this invention requires for the gestalt of the 2nd operation.

[0058] As mentioned above, although the gestalt of operation of this invention was explained, modification in the range which this invention is not limited to the gestalt of this operation, and does not change the summary of invention is possible, and also when it constitutes the manufacture approach of the fullerene of this invention, and its equipment combining the gestalt of each operation, or above mentioned a part or above mentioned all of a modification, it is the right range of this invention. For example, although constituted from two or more circular canal 40a arranged on this alignment to the axial center of a fission reactor 11 in the header tubing 40 with the gestalt of the 5th operation, a clearance may be prepared and two or more straight pipes may be arranged in in the shape of a grid, respectively. Moreover, although the clearance was prepared on this alignment to the axial center of a

fission reactor 11 and two or more 1st header tubing 45 and 2nd header tubing 47 have been arranged with the gestalt of the 6th operation, a clearance may be prepared and the 1st header tubing and the 2nd header tubing may be arranged in the shape of a grid, respectively. Furthermore, although the minor diameter discharge tube 24 of the 2nd burner 14 was produced with heat-resisting steel, such as stainless steel, and the porosity member was produced with the heat-resistant sintered metal with the gestalt of the 3rd and the 4th operation, it is also producible with a cermet and the ceramics.

[0059]

[Effect of the Invention] In the manufacture approach of fullerene according to claim 1 to 3 The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies the coal-for-coke-making-ized hydrogen gasified in the middle of this combustion gas style, and having the 2nd reaction band which makes coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. Since the pressure of the 2nd reaction band is made under into atmospheric pressure, the pyrolysis of coal-for-coke-making-ized hydrogen can advance to homogeneity, the generation effectiveness of fullerene can be raised, and fullerene can be manufactured cheaply and easily in large quantities.

[0060] On the other hand, in the manufacture approach of the fullerene by the above and the well-known combustion method, usual is the same and the fuel for a combustion reaction and the raw material for fullerene generation cannot select a fuel required for a hydrocarbon fuel combustion reaction to arbitration. On the other hand, since the fuel for a combustion reaction and the raw material for manufacture of fullerene can be selected separately according to this invention, when manufacturing fullerene especially on a scale of industry, the cheap original fuel of cost can be freely chosen according to the supply situation of a original fuel.

[0061] It sets to the manufacture approach of fullerene according to claim 2 especially. By being able to keep the conditions of the 2nd reaction band constant over all the cross sections in a furnace, and adjusting the conditions in this band on conditions from which the yield of fullerene serves as max, since the 2nd reaction band is in the downstream of the 1st reaction band Since the field which fullerene generates can be extended to max, compared with the usual combustion method, the yield of fullerene becomes high. On the other hand, although fullerene mainly generates in a flame in the conventional combustion method, generally, a flame has temperature distribution and it is known that fullerene will generate in the specific field of a flame.

[0062] In the manufacture approach of fullerene according to claim 3, since the temperature of the 2nd reaction band is 1000 degrees C or more, the pyrolysis of the supplied coal-for-coke-making-ized hydrogen can be carried out in a short time certainly, and fullerene can be manufactured in large quantities.

[0063] In the manufacturing installation of fullerene according to claim 4 to 13 The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, Since it has the 2nd reaction band which it is [ band ] in the downstream of the 1st reaction band, makes the coal-for-coke-making-ized hydrogen which has the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and was gasified and supplied react in a combustion gas style, and makes fullerene generate Both control of the combustion condition of a fuel and control of the pyrolysis of coal-for-coke-making-ized hydrogen become easy, and it becomes possible to manufacture fullerene in large quantities, cheaply, and easily.

[0064] Especially, in the manufacturing installation of fullerene according to claim 5, since the delivery of the 2nd burner has a clearance in the upstream of the 2nd reaction band, and a large number formation is carried out and it carries out distributed emission of the coal-for-coke-making-ized hydrogen into a combustion gas style, it can pyrolyze coal-for-coke-making-ized hydrogen to homogeneity in combustion gas in a short time, and becomes possible [ making high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen ].

[0065] In the manufacturing installation of fullerene according to claim 6 Since the 2nd burner consists



of a minor diameter discharge tube of a large number arranged by penetrating the 1st reaction band Distributed emission can be carried out uniformly, the coal-for-coke-making-ized hydrogen by which the preheating was carried out into the combustion gas style of the elevated temperature of the 2nd reaction band can be pyrolyzed, and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0066] In the manufacturing installation of fullerene according to claim 7, since the 1st burner emits oxygen content gas and fuel gas independently and it has two or more oxygen content gas nozzles and fuel gas nozzles by which mixture arrangement was carried out, you can carry out diffusive mixing of the oxygen content gas and fuel gas which were supplied, they can make it exist in the 1st reaction band by the uniform mixed state, and become possible [ carrying out the perfect combustion of the fuel gas easily in the 1st reaction band ]. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0067] In the manufacturing installation of fullerene according to claim 8, since it blows off where the head of the 1st burner consisted of a porosity member and oxygen content gas and fuel gas are mixed from a front face, oxygen content gas and fuel gas can be supplied to the 1st reaction band, where premixing is carried out, and it becomes possible to carry out the perfect combustion of the fuel gas easily in the 1st reaction band. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0068] In the manufacturing installation of fullerene according to claim 9, since mixing of oxygen content gas and fuel gas is performed within the 1st burner and oxygen content gas and fuel gas are independently supplied to the 1st burner for another piping, it is not necessary to establish the premixing means of oxygen content gas and fuel gas, and the configuration of the manufacturing installation of fullerene can be simplified.

[0069] In the manufacturing installation of fullerene according to claim 10, since the accumulator which premixing of oxygen content gas and the fuel gas was carried out, and was prepared in the lower part of a head is supplied, structure of the 1st burner can be simplified and the cost of the 1st burner can be reduced.

[0070] In the manufacturing installation of fullerene according to claim 11 the 1st burner Since the oxygen content gas and fuel gas by which have header tubing with which the jet nozzle of many minor diameters set the clearance, and was formed, and premixing was carried out to header tubing are supplied Where premixing is carried out, distributed emission of oxygen content gas and the fuel gas can be carried out in the 1st reaction band, and it becomes possible to carry out the perfect combustion of the fuel gas easily in the 1st reaction band. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0071] In the manufacturing installation of fullerene according to claim 12 The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout oxygen content gas set the clearance, and the 1st burner was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with the 1st header tubing, are arranged, and spout fuel gas set the clearance, and was formed. Since oxygen content gas and fuel gas are independently supplied to the 1st header tubing and the 2nd header tubing for another piping, respectively Diffusive mixing of the oxygen content gas and fuel gas by which distributed emission was carried out can be carried out, they can be in the uniform mixed state, can make it exist in the 1st reaction band, and become possible [ carrying out the perfect combustion of the fuel gas easily in the 1st reaction band ]. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0072] It can prevent that fill up the heat energy consumed when coal-for-coke-making-ized hydrogen pyrolyzed, and the temperature of combustion gas falls since oxygen content gas is mixed in the



manufacturing installation of fullerene according to claim 13 in the coal-for-coke-making-ized hydrogen supplied from the 2nd burner, and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

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**TECHNICAL FIELD**

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[Field of the Invention] This invention relates to the manufacture approach of fullerene, and its equipment.

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PRIOR ART

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[Description of the Prior Art] Fullerene (it may only be hereafter called fullerene) is the generic names of the third carbon allotrope which ranks second to a diamond and a graphite, and it is the carbon molecule of the shape of hollow husks closed in the network of five membered-rings and six membered-rings so that it might be represented by C60 and C70 grade. Although it is comparatively that existence of fullerene was finally checked and it is a comparatively new carbon material, it is admitted that the special molecular structure, therefore specific physical property are shown, for example, innovative application development is being quickly developed over the wide range following fields.

- (1) Application to a superhard ingredient : since purification of the artificial diamond which has a fine crystal grain child by using fullerene as a precursor is possible, use to an abrasion resistance material with added value is expected.
- (2) Application to drugs : research as an application of an anticancer agent, an acquired immunodeficiency syndrome, osteoporosis and the Alzheimer remedy, a contrast medium, a stent ingredient, etc. is advanced by using C60 derivative and an optical device.
- (3) Application to a superconducting material : if metallic potassium is doped to a fullerene thin film, it is discovered that a superconducting material with a high transition temperature called 18K can be made, and since various, attract attention.
- (4) Application to semi-conductor manufacture : it uses that resist structure is further strengthened with mixing C60 with a resist, and the application to next-generation semi-conductor manufacture is expected.

[0003] Also in the fullerene of various carbon numbers, C60 and C70 are comparatively easy to compound, and it is expected that future need so also increases explosively. The approach shown below is mentioned as the manufacture approach of fullerene learned now.

- (1) It is the approach of irradiating the pulse laser of a high energy consistency at the carbon target placed into laser vacuum deposition rare gas, and compounding by evaporation of a carbon atom. The quartz tube with which rare gas flows is placed into an electric furnace, and a graphite sample is placed into the quartz tube. If laser is irradiated and is evaporated in a graphite sample from the upstream of the flow of gas, the soot containing fullerene, such as C60 and C70, will adhere to the wall of the quartz tube with which near the electric furnace outlet got cold. The evaporation per shot of laser is slight and it is unsuitable for extensive manufacture.
- (2) It is the approach to which carry out energization heating and a graphite rod is made to sublime in the container under the reduced pressure filled with resistance heating method gaseous helium. Since the electric resistance loss in a circuit is large, it is unsuitable for extensive manufacture.

[0004] (3) It is the approach to which the carbon of a lifting and an anode plate is made to sublime arc discharge in the condition of having contacted two graphite electrodes lightly in the gaseous helium in number of arc discharge methods 10kPa, or having detached about 1-2mm. It is used for extensive manufacture of the fullerene in a current works scale.

- (4) Instead of using radio frequency heating method resistance heating and arc discharge, it is the approach of heating an eddy current to raw material graphite by RF induction, and evaporating a sink

and raw material graphite.

(5) It is the approach of carrying out the incomplete combustion of the hydrocarbon raw materials, such as benzene, in the mixed gas of inert gas, such as combustion method helium, and oxygen. It is observed as the mass-producing method for being a point usable to liquid fuel etc., and the point that a manufacturing installation is simple, and opposing an arc discharge method in the soot (fullerene etc.) which sub\*\* in that several% of a benzene fuel serves as soot, and the about 10% becomes fullerene although manufacture effectiveness is not good.

(6) It is the approach of carrying out the pyrolysis of the naphthalene thermal decomposition method naphthalene at about 1000 degrees C.

[0005] Thus, although the synthesis method of various fullerene by current is proposed, the method of manufacturing fullerene in large quantities cheaply by any approach until now is not established. A combustion method is considered one of these approaches of the cheapest and efficient manufacture approach, for example, the manufacture approach of the fullerene by burning a carbon inclusion in a flame in the patent reference 1, and collecting condensates in it is indicated. This approach is an approach of manufacturing fullerene by burning a carbon inclusion in a flame, and the fuel for combustion and the raw material of fullerene are the same carbon inclusions substantially. Although fullerene is contained in the soot-like matter and it is generated, a part of this soot-like matter is the so-called carbon black.

[0006] As the manufacture approach of carbon black, the furnace method, a channel process, thermal \*\*, the acetylene method, etc. are learned, and the furnace method is industrially mentioned as the general manufacture approach. The carbon black manufacturing installation (fission reactor) of the shape for example, of a cylinder should be used for this approach, and it should receive \*\*\*\* in the 1st reaction band of the fission reactor concerned. To horizontal or a perpendicular direction, supply oxygen content gas and fuels, such as air, and they are burned. It is made to move to the 2nd reaction band with the cross section which was installed in the lower stream of a river of furnace shaft orientations, and reduced the obtained combustion gas style. It is the approach of supplying coal-for-coke-making-ized hydrogen (stock oil), making it reacting into the gas stream concerned, making carbon black generating, quenching gas by spraying of cooling water etc. to a gas stream further in the 3rd reaction band on the lower stream of a river, and stopping a reaction.

[0007]

[Patent reference 1] \*\*\*\*\* No. 507879 [ six to ] official report

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**EFFECT OF THE INVENTION**

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[Effect of the Invention] In the manufacture approach of fullerene according to claim 1 to 3, The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies the coal-for-coke-making-ized hydrogen gasified in the middle of this combustion gas style, and having the 2nd reaction band which makes coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. Since the pressure of the 2nd reaction band is made under into atmospheric pressure, the pyrolysis of coal-for-coke-making-ized hydrogen can advance to homogeneity, the generation effectiveness of fullerene can be raised, and fullerene can be manufactured cheaply and easily in large quantities.

[0060] On the other hand, in the manufacture approach of the fullerene by the above and the well-known combustion method, usual is the same and the fuel for a combustion reaction and the raw material for fullerene generation cannot select a fuel required for a hydrocarbon fuel combustion reaction to arbitration. On the other hand, since the fuel for a combustion reaction and the raw material for manufacture of fullerene can be selected separately according to this invention, when manufacturing fullerene especially on a scale of industry, the cheap original fuel of cost can be freely chosen according to the supply situation of a original fuel.

[0061] Especially, it sets to the manufacture approach of fullerene according to claim 2, Since the 2nd reaction band is in the downstream of the 1st reaction band and the field which fullerene generates by being able to keep the conditions of the 2nd reaction band constant over all the cross sections in a furnace, and adjusting the conditions in this band on conditions from which the yield of fullerene serves as max can be extended to max, compared with the usual combustion method, the yield of fullerene becomes high. On the other hand, although fullerene mainly generates in a flame in the conventional combustion method, generally, a flame has temperature distribution and it is known that fullerene will generate in the specific field of a flame.

[0062] In the manufacture approach of fullerene according to claim 3, since the temperature of the 2nd reaction band is 1000 degrees C or more, the pyrolysis of the supplied coal-for-coke-making-ized hydrogen can be carried out in a short time certainly, and fullerene can be manufactured in large quantities.

[0063] In the manufacturing installation of fullerene according to claim 4 to 13, The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, Since it has the 2nd reaction band which it is [ band ] in the downstream of the 1st reaction band, makes the coal-for-coke-making-ized hydrogen which has the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and was gasified and supplied react in a combustion gas style, and makes fullerene generate Both control of the combustion condition of a fuel and control of the pyrolysis of coal-for-coke-making-ized hydrogen become easy, and it becomes possible to manufacture fullerene in large quantities, cheaply, and easily.

[0064] Especially, in the manufacturing installation of fullerene according to claim 5, since the delivery

of the 2nd burner has a clearance in the upstream of the 2nd reaction band, and a large number formation is carried out and it carries out distributed emission of the coal-for-coke-making-ized hydrogen into a combustion gas style, it can pyrolyze coal-for-coke-making-ized hydrogen to homogeneity in combustion gas in a short time, and becomes possible [ making high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen ].

[0065] In the manufacturing installation of fullerene according to claim 6, Since the 2nd burner consists of a minor diameter discharge tube of a large number arranged by penetrating the 1st reaction band, distributed emission of it can be carried out uniformly, it can pyrolyze the coal-for-coke-making-ized hydrogen by which the preheating was carried out into the combustion gas style of the elevated temperature of the 2nd reaction band, and becomes possible [ making high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen ].

[0066] In the manufacturing installation of fullerene according to claim 7, since the 1st burner emits oxygen content gas and fuel gas independently and it has two or more oxygen content gas nozzles and fuel gas nozzles by which mixture arrangement was carried out, you can carry out diffusive mixing of the oxygen content gas and fuel gas which were supplied, they can make it exist in the 1st reaction band by the uniform mixed state, and become possible [ carrying out the perfect combustion of the fuel gas easily in the 1st reaction band ]. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0067] In the manufacturing installation of fullerene according to claim 8, since it blows off where the head of the 1st burner consisted of a porosity member and oxygen content gas and fuel gas are mixed from a front face, oxygen content gas and fuel gas can be supplied to the 1st reaction band, where premixing is carried out, and it becomes possible to carry out the perfect combustion of the fuel gas easily in the 1st reaction band. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0068] In the manufacturing installation of fullerene according to claim 9, since mixing of oxygen content gas and fuel gas is performed within the 1st burner and oxygen content gas and fuel gas are independently supplied to the 1st burner for another piping, it is not necessary to establish the premixing means of oxygen content gas and fuel gas, and the configuration of the manufacturing installation of fullerene can be simplified.

[0069] In the manufacturing installation of fullerene according to claim 10, since the accumulator which premixing of oxygen content gas and the fuel gas was carried out, and was prepared in the lower part of a head is supplied, structure of the 1st burner can be simplified and the cost of the 1st burner can be reduced.

[0070] It sets to the manufacturing installation of fullerene according to claim 11, and is the 1st burner, Since the oxygen content gas and fuel gas by which have header tubing with which the jet nozzle of many minor diameters set the clearance, and was formed, and premixing was carried out to header tubing are supplied, where premixing is carried out, distributed emission of oxygen content gas and the fuel gas can be carried out in the 1st reaction band, and it becomes possible to carry out the perfect combustion of the fuel gas easily in the 1st reaction band. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0071] In the manufacturing installation of fullerene according to claim 12, The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout oxygen content gas set the clearance, and the 1st burner was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with the 1st header tubing, are arranged, and spout fuel gas set the clearance, and was formed. Since oxygen content gas and fuel gas are independently supplied to the 1st header tubing and the 2nd header tubing for another piping, respectively Diffusive mixing of the oxygen content gas and fuel gas by which distributed emission was carried out can be carried out, they can be in the uniform mixed state, can make it exist in the 1st

reaction band, and become possible [ carrying out the perfect combustion of the fuel gas easily in the 1st reaction band ]. Consequently, a hot combustion gas style can be formed and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

[0072] It can prevent that fill up the heat energy consumed when coal-for-coke-making-ized hydrogen pyrolyzed, and the temperature of combustion gas falls since oxygen content gas is mixed in the manufacturing installation of fullerene according to claim 13 in the coal-for-coke-making-ized hydrogen supplied from the 2nd burner, and it becomes possible to make high yield of the fullerene made to generate from the pyrolysis object of coal-for-coke-making-ized hydrogen.

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**TECHNICAL PROBLEM**

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[Problem(s) to be Solved by the Invention] However, by the manufacture approach of the above-mentioned usual carbon black, fullerene is hardly generated. In manufacture of fullerene, it has been a big technical problem how the rate of the fullerene contained in the soot-like matter obtained is raised. Generally, manufacture of fullerene is performed under reduced pressure and a diluent may be introduced all over a reaction field. It is known whenever [ these reduced pressure ] that diluent concentration will affect the yield of the above-mentioned fullerene.

[0009] In order to raise the yield of fullerene in the above and the patent reference 1, the approach of supplying energy further is stated to the flame from the external energy source as raising flame temperature and its means. As a desirable energy source, electric resistance heating which heats a flame directly, microwave heating, discharge heating, and counterflow heating that heats a flame by heat exchange with elevated-temperature gas are mentioned.

[0010] By the above and the patent reference 1, pure oxygen is used as an oxidizer for a combustion reaction, and the argon is used as a diluent. This is considered to be effective in gathering the yield of fullerene. However, the amount of the oxygen needed for combustion also becomes extensive, and pure oxygen becomes a special oxygen supply facility is required and expensive [ the manufacturing cost of fullerene ] as a result, when a bomb or supply equipment of dedication etc. tends to be required and it is going to manufacture fullerene on a scale of industry especially.

[0011] So, it has not result in utilization for the reasons of the rate which the volume increases at the time of the actuation under that combustion temperature becomes low since there are many rates of that a flame is not stabilize compared with pure oxygen since the oxygen density is low although it can guess easily use air as an oxidizer of combustion in order to reduce a manufacturing cost in a combustion method, or nitrogen, especially reduced pressure, and passes a nozzle become quick. Since fullerene is various as the exotic material which bears the next generation, and new materials, it is observed, and development of the technique of manufacturing fullerene cheaply and easily in large quantities is desired.

[0012] This invention is made in view of a situation which was mentioned above, and it aims at offering the manufacture approach of the fullerene which manufacture fullerene cheaply and easily in large quantities, and its equipment.

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**MEANS**

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[Means for Solving the Problem] The first reaction band in which this invention persons supply oxygen content gas and a fuel, and burn them in a fission reactor as a result of examining various the optimal combustion methods and manufacturing installations which can manufacture fullerene in large quantities and cheaply, and a hot combustion gas style is made to form, The manufacturing installation of the fullerene which have the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and have the 2nd reaction band which makes coal-for-coke-making-ized hydrogen react and makes fullerene generate is used. Knowledge that fullerene is stably generable in large quantities by maintaining the pressure of the 2nd reaction band under at atmospheric pressure was acquired.

[0014] Namely, the manufacture approach of the fullerene concerning the 1st invention in alignment with said purpose The 1st reaction band which oxygen content gas and a fuel are supplied [ band ], burns them and makes a hot combustion gas style form in a fission reactor, The manufacturing installation of the fullerene characterized by having the coal-for-coke-making-ized hydrogen feed hopper which supplies coal-for-coke-making-ized hydrogen in the middle of this combustion gas style, and having the 2nd reaction band which makes this coal-for-coke-making-ized hydrogen react and makes fullerene generate is used, and the pressure of said 2nd reaction band is made under into atmospheric pressure. Since a fuel and oxygen content gas are supplied and are burned in the 1st reaction band, perfect combustion can be attained easily, for example and a hot combustion gas style can be formed. And by supplying coal-for-coke-making-ized hydrogen into the acquired hot gas stream, the pyrolysis of the coal-for-coke-making-ized hydrogen can be carried out easily, and the generation effectiveness of fullerene can be raised. Moreover, by making the pressure in the 2nd reaction band under into atmospheric pressure, and rarefying the mixed state of coal-for-coke-making-ized hydrogen and combustion gas, the pyrolysis of coal-for-coke-making-ized hydrogen can advance to homogeneity and the generation effectiveness of fullerene can be raised.

[0015] In the manufacture approach of the fullerene concerning the 1st invention, it is desirable that said 2nd reaction band is in the downstream of said 1st reaction band. By establishing the 2nd reaction band in the downstream of the 1st reaction band, the hot combustion gas formed in the 1st reaction band can be immediately introduced into the 2nd reaction band. Consequently, temperature of the 2nd reaction band can be made into an elevated temperature. In the manufacture approach of the fullerene concerning the 1st invention, it is desirable that the temperature of said 2nd reaction band is 1000 degrees C or more. By making temperature of the 2nd reaction band into 1000 degrees C or more, the pyrolysis of the supplied coal-for-coke-making-ized hydrogen can be carried out in a short time certainly.

[0016] The manufacturing installation of the fullerene concerning the 2nd invention in alignment with said purpose The 1st reaction band which oxygen content gas and fuel gas are supplied [ band ] through the 1st burner, burns these, and makes a hot combustion gas style form in a fission reactor, It is in the downstream of this 1st reaction band, and has the 2nd reaction band which makes said coal-for-coke-making-ized hydrogen which has the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen in the style of [ said ] combustion gas, and was gasified and supplied react in said

combustion gas style, and makes fullerene generate. Since combustion of a fuel is performed in the 1st reaction band, control of a combustion condition becomes easy and hot combustion gas can be formed easily. Control of the pyrolysis of coal-for-coke-making-ized hydrogen becomes easy by introducing the obtained hot combustion gas style into the 2nd reaction band, and adjusting gas stream conditions, such as temperature of a hot combustion gas style, the rate of flow, and a flow rate, and the conditions of supply of coal-for-coke-making-ized hydrogen, since the pyrolysis of the coal-for-coke-making-ized hydrogen is supplied and carried out into this hot gas stream.

[0017] As for the delivery of said 2nd burner, in the manufacturing installation of the fullerene concerning the 2nd invention, it is desirable to have a clearance in the upstream of said 2nd reaction band, and for a large number formation to be carried out and to carry out distributed emission of said coal-for-coke-making-ized hydrogen into said combustion gas style. By forming in the upstream of the 2nd reaction band the delivery of the 2nd burner which supplies coal-for-coke-making-ized hydrogen, direct coal-for-coke-making-ized hydrogen can be supplied into the hot combustion gas style which flows from the 1st reaction band, and the pyrolysis of the coal-for-coke-making-ized hydrogen can be carried out easily. Moreover, since distributed emission of the coal-for-coke-making-ized hydrogen is carried out into combustion gas from many deliveries, coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in combustion gas in a short time. As for said 2nd burner, in the manufacturing installation of the fullerene concerning the 2nd invention, it is desirable to consist of a minor diameter discharge tube of a large number arranged by penetrating said 1st reaction band. Since coal-for-coke-making-ized hydrogen is supplied with many minor diameter discharge tubes, distributed emission of the coal-for-coke-making-ized hydrogen can be uniformly carried out into the combustion gas style of the elevated temperature of the 2nd reaction band. Moreover, since a minor diameter discharge tube penetrates the 1st reaction band and is arranged, coal-for-coke-making-ized hydrogen is gradually heated by hot combustion gas, passing through the inside of a minor diameter discharge tube, and can promote the pyrolysis in the inside of the combustion gas style of the elevated temperature of the 2nd reaction band.

[0018] In the manufacturing installation of the fullerene concerning the 2nd invention, mixture arrangement of two or more oxygen content gas nozzles and fuel gas nozzles to which said 1st burner emits independently said oxygen content gas and said fuel gas, respectively may be carried out. Diffusive mixing of the oxygen content gas and fuel gas which were supplied can be carried out, they can be in the uniform mixed state, and can be made to exist in the 1st reaction band by considering as such a configuration. Moreover, in the manufacturing installation of the fullerene concerning the 2nd invention, the head of said 1st burner consists of a porosity member, and can be considered as the configuration which blows off from a front face where said oxygen content gas and said fuel gas are mixed. By considering as such a configuration, oxygen content gas and fuel gas can be supplied to the 1st reaction band, where premixing is carried out.

[0019] In the manufacturing installation of the fullerene concerning the 2nd invention, mixing of said oxygen content gas and said fuel gas is performed within said 1st burner, and it can consider as the configuration to which said oxygen content gas and said fuel gas are independently supplied for another piping at said 1st burner. Since mixing of oxygen content gas and fuel gas is performed within the 1st burner, it is not necessary to establish separately the premixing means of oxygen content gas and fuel gas, and the configuration of the manufacturing installation of fullerene becomes easy. In the manufacturing installation of the fullerene concerning the 2nd invention, said oxygen content gas and said fuel gas can be considered as the configuration supplied to the accumulator which premixing was carried out and was prepared in the lower part of said head. Since premixing of oxygen content gas and the fuel gas is carried out and they are supplied to the accumulator of the lower part of a head, structure of the 1st burner can be simplified.

[0020] In the manufacturing installation of the fullerene concerning the 2nd invention, said 1st burner has header tubing with which the jet nozzle of many minor diameters set the clearance, and was formed, and can consider it as the configuration to which said oxygen content gas with which premixing was carried out to this header tubing, and said fuel gas are supplied. By considering as such a configuration,

where premixing is carried out, distributed emission of oxygen content gas and the fuel gas can be carried out in the 1st reaction band. In the manufacturing installation of the fullerene concerning the 2nd invention said 1st burner The 1st header tubing with which the jet nozzle of the minor diameter of a large number which spout said oxygen content gas set the clearance, and was formed, It has the 2nd header tubing with which the jet nozzle of the minor diameter of a large number which have a clearance with said 1st header tubing, are arranged, and spout said fuel gas set the clearance, and was formed. It can consider as the configuration to which said oxygen content gas and said fuel gas are independently supplied for another piping, respectively at said 1st header tubing and said 2nd header tubing. Diffusive mixing of the oxygen content gas and fuel gas by which distributed emission was carried out can be carried out, they can be in the uniform mixed state, and can be made to exist in the 1st reaction band by considering as such a configuration.

[0021] In the manufacturing installation of the fullerene concerning the 2nd invention, oxygen content gas is mixable in the coal-for-coke-making-ized hydrogen supplied from said 2nd burner. In the pyrolysis of coal-for-coke-making-ized hydrogen, the temperature of combustion gas falls by the pyrolysis of coal-for-coke-making-ized hydrogen for endothermic reaction. For this reason, it can prevent that fill up the heat energy consumed when a part of coal-for-coke-making-ized hydrogen was burned in the 2nd reaction band, heat energy was generated and coal-for-coke-making-ized hydrogen pyrolyzed by mixing oxygen content gas in coal-for-coke-making-ized hydrogen, and the temperature of combustion gas falls.

[0022]

[Embodiment of the Invention] Then, referring to the attached drawing, it explains per gestalt of the operation which materialized this invention, and an understanding of this invention is presented. The explanatory view of the fullerene manufacturing installation which applied the manufacture approach of the fullerene which drawing 1 (A) and (B) require for the gestalt of operation of the 1st of this invention, respectively here, The explanatory view of the manufacturing installation of the fullerene which a plane section Fig., drawing 2 (A), and (B) require for the gestalt of operation of the 2nd of this invention, respectively, The explanatory view of the manufacturing installation of the fullerene which a plane section Fig., drawing 3 (A), and (B) require for the gestalt of operation of the 3rd of this invention, respectively, A plane section Fig., the partial explanatory view of the manufacturing installation of the fullerene which drawing 4 requires for the gestalt of operation of the 4th of this invention, The explanatory view of the manufacturing installation of the fullerene which drawing 5 (A) and (B) require for the gestalt of operation of the 5th of this invention, respectively, a plane section Fig., drawing 6 (A), and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 6th of this invention, and a plane section Fig., respectively.

[0023] The manufacture approach of the fullerene concerning the gestalt of operation of the 1st of this invention is explained using drawing 1. The manufacture approach of the fullerene concerning the gestalt of the 1st operation is related with the approach of introducing coal-for-coke-making-ized hydrogen into the manufacturing installation 3 of the fullerene constituted by forming the 1st reaction band 1 and the 2nd reaction band 2 in fission reactor 3a, and manufacturing fullerene by burning.

[0024] The manufacturing installation 3 of fullerene has the 2nd reaction band 2 which coal-for-coke-making-ized hydrogen is supplied [ band ], makes it react the 1st reaction band 1 in which a combustion gas style is made to form, and in the style of [ which were formed there ] combustion gas, and makes fullerene generate. The 2nd reaction band 2 may be in the downstream of the direction of a combustion gas style (it may be hereafter called "shaft orientations") which may be the almost same field (an outside or inside) as the 1st reaction band 1, and was formed in the 1st reaction band 1.

[0025] Drawing 1 shows the case where the 2nd reaction band 2 is located on the lower stream of a river of the 1st reaction band 1.

Generally in the [1st reaction band] 1st reaction band 1, a combustion gas style hot by supplying a fuel and oxygen content gas and burning them, respectively is generated toward the lower stream of a river of the 2nd reaction band 2, i.e., fission reactor 3a, from a fuel feed hopper and oxygen content gas supply opening.

[0026] Even if supply of a fuel and oxygen content gas is the so-called premixing method mixed before entering in fission reactor 3a, it may be the so-called diffusive-mixing method supplied to fission reactor 3a from the nozzle which became independent, respectively. In drawing 1, in the case of a diffusive-mixing method, a fuel is supplied from the central fuel feed hopper 7, and it supplies oxygen content gas from the oxygen content gas supply openings 5 and 6 of the perimeter. Moreover, a premixing method and a diffusive-mixing method may be combined, for example, in drawing 1, from the oxygen content gas supply opening 5, what mixed oxygen content gas with the fuel beforehand may be supplied, and the fuel from the fuel feed hopper 7 may be independently supplied for oxygen content gas from the oxygen content gas supply opening 6, respectively.

[0027] It may be the purpose that this 1st reaction band 1 generates hot combustion gas, and that combustion method may be what kind of well-known combustion methods, such as premixed combustion, diffusive burning, laminar-flow combustion, turbulent flow combustion, and elevated-temperature air combustion. Moreover, although combustion in the 1st reaction band 1 may be perfect combustion or you may be incomplete combustion as long as the temperature which becomes generable [ fullerene ] in the 2nd reaction band 2 is acquired, it is desirable that it is perfect combustion with the large calorific value to fuel used. When the 1st reaction band 1 is incomplete combustion with the so-called superfluous fuel, the soot-like matter which contains fullerene even in the 1st reaction band 1 may generate.

[0028] However, the combustion by the lean mixture whose oxygen required for combustion is more than the amount of stoichiometry oxygen of the combustion in this 1st reaction band 1 is preferably better. As oxygen content gas, the gas which mixed non-flammable gas, such as argon gas and nitrogen gas, at a rate of arbitration can be used for air, oxygen gas, or these. NOX especially in elevated-temperature combustion Pure oxygen may be used in order to suppress generating. In order to gather the yield of fullerene, it is desirable to dilute using rare gas etc. in a combustion process. Rare gas may be supplied from the exclusive nozzle for supply, and may be beforehand mixed in a fuel, coal-for-coke-making-ized hydrogen, and oxygen content gas.

[0029] As a fuel, coal system liquid fuel, such as petroleum system liquid fuel, such as fuel gas, such as hydrogen, a carbon monoxide, natural gas, and petroleum gas, a fuel oil, benzene, and toluene, and creosote oil, can be used. Especially, as a fuel used with the gestalt of this operation, fuel gas is desirable. Moreover, although what is necessary is for fullerene to obtain just to adjust suitably the mean temperature in the 1st reaction band 1 at the time of fullerene manufacture, it is preferably made into 1600 degrees C or more still more preferably 1300 degrees C or more. This is because the productivity of fullerene goes up, so that the temperature of combustion gas is an elevated temperature. Even if an upper limit is too high not much, the productivity of fullerene may fall. Moreover, what is necessary is just to determine after taking into consideration the heat-resistant problem by the quality of the material of a fission reactor.

[0030] If opening of the arrangement of the fuel feed hopper 7 and the oxygen content gas supply openings 5 and 6 is carried out to fission reactor 3a, it is arbitrary. In drawing 1, opening of the fuel feed hopper 7 and the oxygen content gas supply openings 5 and 6 is carried out to the same fission reactor 3a side. The configuration of each feed hoppers 5, 6, and 7 which are carrying out opening into fission reactor 3a may be arbitrary, and may be the indeterminate form of the shape of a polygon, such as an approximate circle form, an ellipse form, and the shape of a trigonum and a rectangular head, a gourd mold, etc.

[0031] As for fission reactor 3a internal pressure, it is desirable that it is under atmospheric pressure, and the more desirable range is 10 - 300torr.

Coal-for-coke-making-ized hydrogen is supplied from the coal-for-coke-making-ized hydrogen feed hopper 4 in the style of [ which was formed in the 1st reaction band 1 ] combustion gas, and fullerene is made to generate in the [2nd reaction band] 2nd reaction band 2 by carrying out partial combustion of a part of this coal-for-coke-making-ized hydrogen. In order to carry out partial combustion, it is good also considering the combustion in the 1st reaction band 1 as hyperoxia so that oxygen may remain.

Moreover, a nozzle may be arranged to the 2nd reaction field 2, and oxygen content gas may be supplied

to it from an oxygen content gas supply nozzle.

[0032] Under the present circumstances, as for the above-mentioned coal-for-coke-making-ized hydrogen supplied into combustion gas, or oxygen content gas, it is desirable to be supplied in fission reactor 3a as much as possible at homogeneity. For this reason, it is desirable to be equally arranged so well that many by the number of the coal-for-coke-making-ized hydrogen feed hopper 4 installed in the 2nd reaction band 2 and an oxygen content gas supply nozzle in fission reactor 3a.

[0033] What is necessary is just to choose the die length of the 2nd reaction band suitably according to the magnitude of fission reactor 3a, the class of fullerene to manufacture, etc. The location and configuration of the 2nd reaction band may be arbitrary, and may be the inside of the 1st reaction band, or may be an outside, and as shown in drawing 1, they may be in the downstream of the 1st reaction band 1. It is more desirable for the cross-section configuration of the 2nd reaction band not to change, although the configuration of the 2nd reaction band is also arbitrary. The reason is that it will have effect which is not desirable on the fullerene to generate if influenced by the flow by the cross-section configuration of the 2nd reaction band changing in the process which fullerene generates of turbulence.

[0034] Although what is necessary is just to choose the mean temperature of the 2nd reaction band 2 suitably by the fullerene to manufacture, in order that coal-for-coke-making-ized hydrogen may evaporate and react to homogeneity, it is desirable that it is an elevated-temperature ambient atmosphere enough. It is desirable that it is specifically 1000 degrees C or more, and it is especially desirable that it is 1700-1900 degrees C 1000-1900 degrees C especially. Moreover, in the 2nd reaction band 2, it is desirable to control the oxygen density in combustion gas as much as possible. It is because there is a thing of coal-for-coke-making-ized hydrogen, the generation reaction band 2, i.e., 2nd reaction band, of fullerene, which combustion takes place actively in part, therefore the ununiformity of the temperature in the 2nd reaction band 2 produces when oxygen exists so much in combustion gas. the oxygen density in combustion gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably.

[0035] In the gestalt of this operation, the location which supplies coal-for-coke-making-ized hydrogen is arbitrary and can prepare a coal-for-coke-making-ized hydrogen feed hopper according to the configuration of a fission reactor. For example, a coal-for-coke-making-ized hydrogen feed hopper may be prepared in the contraction section which may prepare a coal-for-coke-making-ized hydrogen feed hopper in the part from which the path of fission reactor 3a serves as max, and the path is reducing. Furthermore, as it \*\* to drawing 1, the coal-for-coke-making-ized hydrogen feed hopper 4 may be formed in the contraction section which the part from which the path of fission reactor 3a serves as max, and the path are reducing, respectively. The rate of flow of the gas in the location where coal-for-coke-making-ized hydrogen is introduced, the strength of a turbulent flow, etc. are controllable by the location of the coal-for-coke-making-ized hydrogen feed hopper 4.

[0036] As coal-for-coke-making-ized hydrogen, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of a raw material uses an aromatic series system hydrocarbon especially.

[0037] Two or more locations of the coal-for-coke-making-ized hydrogen feed hopper in a fission reactor may be prepared on the cross-section periphery of the flow direction of combustion gas, and the location which has two or more coal-for-coke-making-ized hydrogen feed hoppers on still such same periphery may be established in the flow direction of combustion gas multistage. In order to make generation reaction time of fullerene into homogeneity and for physical properties to obtain uniform fullerene, it is desirable to install as many coal-for-coke-making-ized hydrogen feed hoppers as possible on the same periphery.

[0038] Moreover, although the form of the nozzle used for the coal-for-coke-making-ized hydrogen feed hopper 4 can be chosen suitably, when using the coal-for-coke-making-ized hydrogen of a liquid, in order to spray on homogeneity minutely more, it is desirable that the diameter of an initial drop of the coal-for-coke-making-ized hydrogen immediately after spraying from nozzles, such as 2 hydraulic nozzles which inject the supplied liquid with another liquid, considers as a small thing as much as possible. Although what is necessary is just to choose suitably, before the coal-for-coke-making-ized hydrogen sprayed on the 2nd reaction band 2 evaporates, as for the coal-for-coke-making-ized hydrogen supply approaches, such as a diameter of opening of the coal-for-coke-making-ized hydrogen feed hopper 4, a form, protrusion condition into a furnace, a supply include angle to a combustion gas style, and a gas-liquid ratio, the rate of flow, a flow rate, temperature, etc., it is desirable to spray on conditions which do not adhere to the furnace wall of the 2nd reaction band 2. By spraying such, the foreign matter in the soot-like matter obtained can be reduced.

[0039] The thing of arbitration can be used if it is the quality of the material which has thermal resistance, such as a metal and refractories, as internal insulation which constitutes the 1st reaction band 1 and the 2nd reaction band 2. Since the temperature of internal combustion gas becomes beyond metaled heat-resistant temperature when using a metal, it is necessary to cool from the outside by taking structures, such as rolling water cooled jacket structure and a water-cooled tube. As ingredients other than a metal, there are SiC, a diamond, nitriding aluminum, silicon nitride, ceramic system refractory material, etc., for example.

[0040] It is made into the structure which cools preferably 1000 degrees C or less of combustion gas styles containing the soot-like matter (the thing in the middle of a reaction is included) containing fullerene at 800 degrees C or less from the 2nd reaction band 2 after the downstream. Water etc. may be sprayed from a reaction halt fluid feed hopper, and, specifically, you may cool by passing the passage which cooled the exterior according to water-cooled structure etc. Especially, especially when the path of passage is small, even if it does not consider as water-cooled structure, it may fully be cooled by the natural heat dissipation to atmospheric air.

[0041] It dissociates with gas (not shown) and the fullerene and the soot-like matter which were cooled are recovered by the uptake bag filter prepared in the point of passage. The extraction approach of fullerene can use well-known general processes, such as making it adhere to such a bag filter or a passage wall etc.

[0042] As shown in drawing 2, the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 2nd of this invention The 1st reaction band 13 which the oxygen content gas and fuel gas which were supplied through the 1st burner 12 in the fission reactor 11 burn, and forms a hot combustion gas style, It is in the downstream of the 1st reaction band 13, and has the 2nd reaction band 16 which makes the coal-for-coke-making-ized hydrogen which has the delivery 15 of the 2nd burner 14 which supplies coal-for-coke-making-ized hydrogen in the style of combustion gas, and was gasified and supplied react in a combustion gas style, and makes fullerene generate. Hereafter, these are explained to a detail. The fission reactor 11 is equipped with the cylindrical shape-like side-attachment-wall section 17 and the edge wall 19 which it connects with the end side of the side-attachment-wall section 17, and an outer diameter contracts gradually, and forms the exhaust port 18. The side-attachment-wall section 17 and the edge wall 19 consist of heat-resisting steel, such as stainless steel. Furthermore, the refractories which are not illustrated are lined by the inner skin by the side of the other end of the side-attachment-wall section 17. As refractories, the refractory brick of the quality of an alumina and the unshaped refractories of the quality of an alumina can be used, for example. Moreover, the end side of the exhaust pipe which is not illustrated is connected to an exhaust port 18, and the other end side of an exhaust pipe is connected to the exhaust air pump. For this reason, while changing the inside of a fission reactor 11 into the reduced pressure condition of under atmospheric pressure, the combustion gas containing the soot-like matter generated in the fission reactor 11 can be discharged outside from the inside of a fission reactor 11.

[0043] The 1st burner 12 attached in base 17a by the side of the other end of the side-attachment-wall section 17 has two or more oxygen content gas nozzles 21 linked to the oxygen content gas supply

piping 20, and the fuel gas nozzle 23 linked to the fuel gas charging line 22, and mixture arrangement of each of these gas nozzles 21 and 23 is carried out at base 17a. Moreover, the oxygen content gas nozzle 21 and the fuel gas nozzle 23 are formed with heat-resisting steel, such as stainless steel. For this reason, after the oxygen content gas supplied from the oxygen content gas nozzle 21 and the fuel gas supplied from the fuel gas nozzle 23 are emitted, diffusive mixing of it will be carried out, it will be in the uniform mixed state, and burns in the 1st reaction band 13. And the formed hot combustion gas style flows into the 2nd reaction band 16 of the downstream. The 2nd burner 14 attached in the other end side of the side-attachment-wall section 17 consists of a minor diameter discharge tube 24 (for example, formed with heat-resisting steel, such as stainless steel) of a large number arranged by penetrating the 1st reaction band 13. Consequently, the delivery 15 established in the tip side of the minor diameter discharge tube 24 has a clearance in the upstream of the 2nd reaction band 16, and is arranged at it. Moreover, the end face side of each minor diameter discharge tube 24 is connected to the coal-for-coke-making-ized hydrogen charging line 25. For this reason, direct coal-for-coke-making-ized hydrogen can be supplied at homogeneity into the hot combustion gas style which flows from the 1st reaction band 13, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time.

[0044] Next, the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene concerning the gestalt of operation of the 2nd of this invention is explained to a detail. The fuel gas nozzle 23 to fuel gas is supplied for oxygen content gas from the oxygen content gas nozzle 21, a combustion gas style hot by burning these is formed, and it is made to circulate toward the lower stream of a river of a fission reactor 11. As oxygen content gas, the gas (for example, the concentration of inert gas can be adjusted in not more than 90 mol % exceeding 0 or 0) which mixed inert gas, such as argon gas, at a rate of arbitration can be used for the oxygen gas which is a source of oxygen. As a source of oxygen, from a viewpoint of the yield of fullerene, oxygen gas is desirable and air is desirable from a viewpoint of the ease of carrying out of acquisition of the source of oxygen etc. In order to raise especially combustion temperature, before these oxygen content gas is supplied in a fission reactor 11, it is desirable to become hot beforehand. As the approach of a preheating, what kind of well-known approaches, such as heat exchange with the combustion gas which used the heat exchanger, and the so-called regeneration burner, may be used. With [ the temperature of this preheating ] ordinary temperature [ beyond ], what kind of temperature is sufficient, but in order to gather the yield of fullerene, the high temperature is more desirable as much as possible. It is desirable more preferably that it is beyond the self-ignition temperature of combustion gas.

[0045] What gasified coal system liquid fuel which gasified petroleum system liquid fuel, such as fuel gas, such as a carbon monoxide, natural gas, and petroleum gas, and a fuel oil, such as a thing and creosote oil, as fuel gas can be used. Fuel gas, such as natural gas and petroleum gas, is desirable especially. Moreover, in order to gather the yield of fullerene, it is desirable to also dilute fuel gas using inert gas etc.

[0046] Then, the combustion gas style which fuel gas burns and forms under oxygen content gas is explained. While adjusting the amount of the fuel gas supplied from the fuel gas nozzle 23 on the conditions which fuel gas burns completely, and the amount of oxygen gas supplied from the oxygen content gas nozzle 21 and supplying the 1st reaction band 13, combustion of fuel gas starts with an ignition means to by\_which hold the inside of a fission reactor 11 and an exhaust-air pump does not illustrate it in the condition of 10 - 300torr more preferably under atmospheric pressure through the exhaust pipe which was connected to the exhaust port 18 and which is not illustrated. Here, fuel gas and oxygen content gas become independent respectively, and since it is emitted in the 1st reaction band 13 from the oxygen content gas nozzle 21 which separated distance and was distributed, and the fuel gas nozzle 23, they can make homogeneity the combustion condition in the 1st reaction band 13. Moreover, since the pressure in a fission reactor 11 has become under atmospheric pressure in addition to diluting with inert gas, such as argon gas, and falling, the oxygen gas concentration in oxygen content gas can change the combustion condition in the 1st reaction band 13 into the condition that it was similar with the elevated-temperature air combustion condition. Consequently, combustion of fuel gas advances to homogeneity and can make temperature of the 1st reaction band 13 homogeneity and an elevated



temperature (for example, 1000-1900 degrees C, preferably 1700-1900 degrees C).

[0047] Since the hot combustion gas formed in the 2nd reaction band 16 in the 1st reaction band 13 flows, the temperature of the upstream of the 2nd reaction band 16 becomes a 1000-1900-degree C elevated temperature. Distributed emission of the coal-for-coke-making-ized hydrogen is carried out into the combustion gas style of the upstream of the 2nd reaction band 16 from each delivery 15 of the minor diameter discharge tube 24 of a large number arranged by penetrating the 1st reaction band 13. Here, since the 1st reaction band 13 is penetrated and it is arranged, since the preheating is carried out while passing through the inside of the minor diameter discharge tube 24, the minor diameter discharge tube 24 pyrolyzes coal-for-coke-making-ized hydrogen, shortly after being emitted into a hot combustion gas style from a delivery 15. Consequently, the high pyrolysate of labile exists in combustion gas, and a fullerene precursor is formed when these coalesce. And it grows up, while a fullerene precursor moves with a combustion gas style, and it becomes fullerene. In addition, since the pyrolysis of coal-for-coke-making-ized hydrogen is endothermic reaction, heat energy is taken from combustion gas and the temperature of combustion gas falls. For this reason, oxygen content gas is mixed in coal-for-coke-making-ized hydrogen, a part of raw material carbon hydrogen is burned, and you may make it supply heat energy. however, a part of raw material carbon hydrogen -- since the ununiformity of the temperature in the 2nd reaction band 16 will arise and the generation effectiveness of fullerene will fall, if combustion takes place actively -- the oxygen density in combustion gas -- desirable -- less than [ 3vol% ] -- it is 0.05 - 1vol% still more preferably.

[0048] As raw material carbon hydrogen, the thing of well-known arbitration can be used conventionally. For example, aromatic series system hydrocarbons, such as benzene, toluene, a xylene, naphthalene, and an anthracene, Coal system hydrocarbons, such as creosote oil and a carboxylic-acid oil, ethylene heavy-ends oil, Aliphatic saturated hydrocarbon, such as petroleum system heavy oil, such as FCC oil (fluidized-catalytic-cracking residue oil), acetylene series unsaturated hydrocarbon, the hydrocarbon of ethylene series, a pentane, and a hexane, etc. is mentioned, and these may be mixed and used at a rate of independent or arbitration. It is desirable to use the aromatic series system hydrocarbon refined especially, and aromatic series system hydrocarbons, such as benzene and toluene, are especially desirable. Its higher one is desirable, and it is so good that its purity is close to 100% in case the purity of the raw material carbon hydrogen which mainly serves as a raw material uses an aromatic series system hydrocarbon especially.

[0049] As shown in drawing 3 , it is the description that premixing of oxygen content gas and the fuel gas is carried out, and the manufacturing installation 26 of the fullerene concerning the gestalt of operation of the 3rd of this invention is supplied to the 1st burner 27. Therefore, only the 1st burner 27 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. It is produced with the heat-resistant metal and the 1st burner 27 has the head 28 which the whole surface side has exposed to the 1st reaction band 13 of a fission reactor 11, and the accumulator 29 prepared in the lower part of a head 28. And each minor diameter discharge tube 24 of the 2nd burner 14 opened the predetermined clearance mutually, penetrated the accumulator 29 and the head 28 from the lower part of an accumulator 29, and has projected them in the fission reactor 11.

[0050] Here, the head 28 consists of porosity members of sintering metal. If the porosity member has structure equipped with many free passage holes which are open for free passage to a side on the other hand from the whole surface side, it considers as the mixed gas which carried out premixing of oxygen content gas and the fuel gas to the accumulator 29 prepared in the lower part of a head 28 and it supplies from the mixed-gas charging line 30 Mixed gas can be moved to the field exposed to the 1st reaction band 13 side from the field by the side of an accumulator 29 through the free passage hole in a head 28, and can be spouted in the 1st reaction band 13. Therefore, combustion gas hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be



pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing facility 26 of the fullerene concerning the gestalt of operation of the 3rd of this invention requires for the gestalt of the 2nd operation.

[0051] In the manufacturing installation 31 of the fullerene concerning the gestalt of operation of the 4th of this invention, since oxygen content gas and fuel gas are independently supplied to the 1st burner 32 for another piping, it is the description that the manufacturing installation 26 of the fullerene concerning the gestalt of the 3rd operation differs from the structure of the 1st burner 32. Therefore, only the 1st burner 32 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. That is, as shown in drawing 4, the 1st burner 32 is produced with a heat-resistant metal, and has two or more gas blenders 35 which have an exhaust nozzle in the head 33 which consists of a porosity member of sintering metallicity which has a free passage hole, the accumulator 34 prepared in the lower part of a head 33, and an accumulator 34. And each minor diameter discharge tube 24 of the 2nd burner 14 opened the predetermined clearance mutually, penetrated the accumulator 34 and the head 33 from the lower part of an accumulator 34, and has projected them in the fission reactor 11. Moreover, the aspirator-type mixer which attracts oxygen content gas and is mixed by the flow of fuel gas as a gas blender 35 can be used.

[0052] If oxygen content gas and fuel gas are independently supplied to each gas blender 35 by considering as such a configuration by the oxygen content gas supply piping 36 and the fuel gas charging line 37, respectively, oxygen content gas and fuel gas will flow in an accumulator 34 as mixed gas from the exhaust nozzle of a gas blender 35, being mixed. And the mixed gas which flowed in the accumulator 34 can be moved to the field exposed to the 1st reaction band 13 side from the field by the side of an accumulator 34 through the free passage hole in a head 33, and can be spouted in the 1st reaction band 13. Therefore, a combustion gas style hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time.

[0053] In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 26 of the fullerene which the manufacture approach of the fullerene which used the manufacturing facility 31 of the fullerene concerning the gestalt of operation of the 4th of this invention requires for the gestalt of the 3rd operation.

[0054] It be the description that the 1st burner 41 which have the header tubing 40 with which the jet nozzle 39 of the minor diameter of a large number which the mixed gas to which the manufacturing installation 38 of the fullerene which start the gestalt of operation of the 5th of this invention as show in drawing 5 be attached in base 17a by the side of the other end of the side attachment wall section 17, and premixing of oxygen content gas and the fuel gas be carried out spout set a clearance, and be form be supply. Therefore, only the 1st burner 41 with which structures differ is explained, the same sign is given to the same component as the manufacturing facility 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted.

[0055] The header tubing 40 had two or more circular canal 40a which prepared the clearance on this alignment, respectively and has been arranged to the axial center of a fission reactor 11, and has connected each circular canal 40a to mixed-gas charging line 30a. And through the clearance between each circular canal 40a, each minor diameter discharge tube 24 of the 2nd burner 14 penetrates the 1st reaction band 13, and is arranged. Therefore, if the mixed gas which carried out premixing of oxygen content gas and the fuel gas is supplied to each circular canal 40a through mixed-gas charging line 30a, mixed gas will be spouted in the 1st reaction band 13 from each jet nozzle 39 of each circular canal 40a.

For this reason, a combustion gas style hot in the 1st reaction band 13 can be formed by burning the mixed gas which blew off in the 1st reaction band 13. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing installation 38 of the fullerene concerning the gestalt of operation of the 5th of this invention requires for the gestalt of the 2nd operation.

[0056] As compared with the manufacturing installation 10 of the fullerene which the manufacturing installation 42 of the fullerene concerning the gestalt of operation of the 6th of this invention requires for the gestalt of operation of the 2nd of this invention, it is the description that the structures of the 1st burner 43 differ. Therefore, only the 1st burner 43 with which structures differ is explained, the same sign is given to the same component as the manufacturing installation 10 of the fullerene concerning the gestalt of the 2nd operation, and detailed explanation is omitted. Namely, as shown in drawing 6, the 1st burner 43 attached in base 17a by the side of the other end of the side-attachment-wall section 17 is produced with a heat-resistant metal. It has the 2nd header tubing 47 with which the jet nozzle 46 of the minor diameter of a large number which the 1st header tubing 45 with which the jet nozzle 44 of the minor diameter of a large number which spout oxygen content gas set the clearance, and was formed, and the 1st header tubing 45 have a clearance, are arranged, and spout fuel gas set the clearance, and was formed. Furthermore, the oxygen content gas supply piping 20 and the fuel gas charging line 22 which supply independently oxygen content gas and said fuel gas, respectively are connected to the 1st header tubing 45 and the 2nd header tubing 47. Moreover, each minor diameter discharge tube 24 of the 2nd burner 14 penetrated base 17a through the clearance between the 1st header tubing 45 and the 2nd header tubing 47, and has projected it in the fission reactor 11.

[0057] Oxygen content gas can be supplied to the 1st header 45 through the oxygen content gas supply piping 20, and it can be made to blow off from the jet nozzle 44 in a fission reactor 11 by considering as such a configuration. Moreover, fuel gas can be supplied to the 2nd header 47 through the fuel gas charging line 22, and it can be made to blow off from the jet nozzle 46 in a fission reactor 11. After the oxygen content gas and fuel gas which blew off from each jet nozzles 44 and 46 are emitted, diffusive mixing of them will be carried out, they will be in the uniform mixed state, and burn in the 1st reaction band 13. And the formed hot combustion gas flows into the 2nd reaction band 16 of the downstream. And the coal-for-coke-making-ized hydrogen supplied through the coal-for-coke-making-ized hydrogen charging line 25 into the flowing hot combustion gas style from the 1st reaction band 13 can be supplied from the delivery 15 of each minor diameter discharge tube 24, and coal-for-coke-making-ized hydrogen can be pyrolyzed to homogeneity in a short time. In addition, since it is substantially the same, detailed explanation is abbreviated to the manufacture approach of the fullerene which used the manufacturing installation 10 of the fullerene which the manufacture approach of the fullerene which used the manufacturing installation 42 of the fullerene concerning the gestalt of operation of the 6th of this invention requires for the gestalt of the 2nd operation.

[0058] As mentioned above, although the gestalt of operation of this invention was explained, modification in the range which this invention is not limited to the gestalt of this operation, and does not change the summary of invention is possible, and also when it constitutes the manufacture approach of the fullerene of this invention, and its equipment combining the gestalt of each operation, or above mentioned a part or above mentioned all of a modification, it is the right range of this invention. For example, although constituted from two or more circular canal 40a arranged on this alignment to the axial center of a fission reactor 11 in the header tubing 40 with the gestalt of the 5th operation, a clearance may be prepared and two or more straight pipes may be arranged in the shape of a grid, respectively. Moreover, although the clearance was prepared on this alignment to the axial center of a fission reactor 11 and two or more 1st header tubing 45 and 2nd header tubing 47 have been arranged

with the gestalt of the 6th operation, a clearance may be prepared and the 1st header tubing and the 2nd header tubing may be arranged in in the shape of a grid, respectively. Furthermore, although the minor diameter discharge tube 24 of the 2nd burner 14 was produced with heat-resisting steel, such as stainless steel, and the porosity member was produced with the heat-resistant sintered metal with the gestalt of the 3rd and the 4th operation, it is also producible with a cermet and the ceramics.

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[Translation done.]

**\* NOTICES \***

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1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

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**DESCRIPTION OF DRAWINGS**

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**[Brief Description of the Drawings]**

[Drawing 1] (A) and (B) are the explanatory view of the fullerene manufacturing installation which applied the manufacture approach of the fullerene concerning the gestalt of operation of the 1st of this invention, and a plane section Fig., respectively.

[Drawing 2] (A) and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 2nd of this invention, and a plane section Fig., respectively.

[Drawing 3] (A) and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 3rd of this invention, and a plane section Fig., respectively.

[Drawing 4] It is the partial explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 4th of this invention.

[Drawing 5] (A) and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 5th of this invention, and a plane section Fig., respectively.

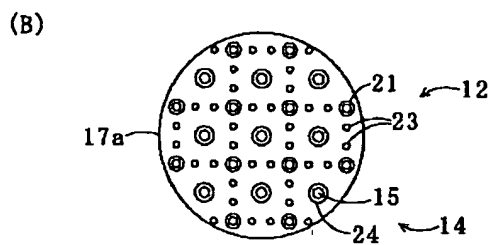
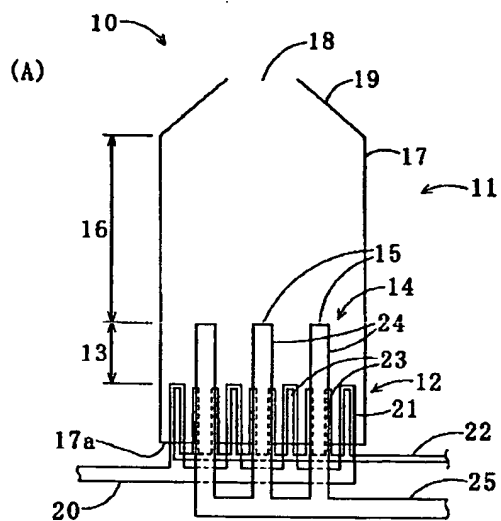
[Drawing 6] (A) and (B) are the explanatory view of the manufacturing installation of the fullerene concerning the gestalt of operation of the 6th of this invention, and a plane section Fig., respectively.

**[Description of Notations]**

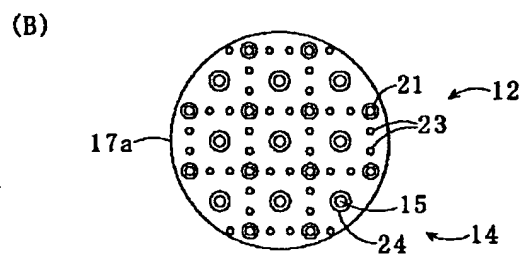
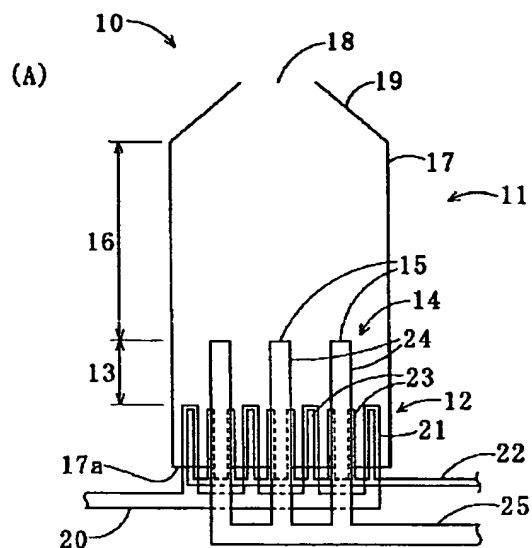
The 1st reaction band, the 2:2nd reaction band, 3 : 1: The manufacturing installation of fullerene, 3a : A fission reactor, 4: coal-for-coke-making-ized hydrogen feed hopper, 5, 6: oxygen content gas supply opening, 7: The manufacturing installation of a fuel feed hopper and 10: fullerene, 11: fission reactor, 12 : The 1st burner, 13: The 1st reaction band, 14 : The 2nd burner, 15: delivery, the 16: 2nd reaction band, 17: The side-attachment-wall section, a 17a: base, 18: exhaust port, 19: edge wall, 20 : Oxygen content gas supply piping, 21: An oxygen content gas nozzle, 22: fuel gas charging line, 23 : A fuel gas nozzle, 24: A minor diameter discharge tube, 25: coal-for-coke-making-ized hydrogen charging line, 26 : The manufacturing installation of fullerene, 27 : A 1st burner, 28: head, 29: accumulator, 30, and 30a: mixed-gas charging line, 31: The manufacturing installation of fullerene, 32 : The 1st burner, 33: head, 34 : An accumulator, 35: gas blender, 36: oxygen content gas supply piping, 37: fuel gas charging line, the manufacturing installation of 38: fullerene, 39: jet nozzle, 40: header tubing, a 40a: circular canal, and 41: - the manufacturing installation of the 1st burner and 42: fullerene, and 43: -- the 1st burner, 44: jet nozzle, and 45: -- the 1st header tubing, 46: jet nozzle, and 47: -- the 2nd header tubing

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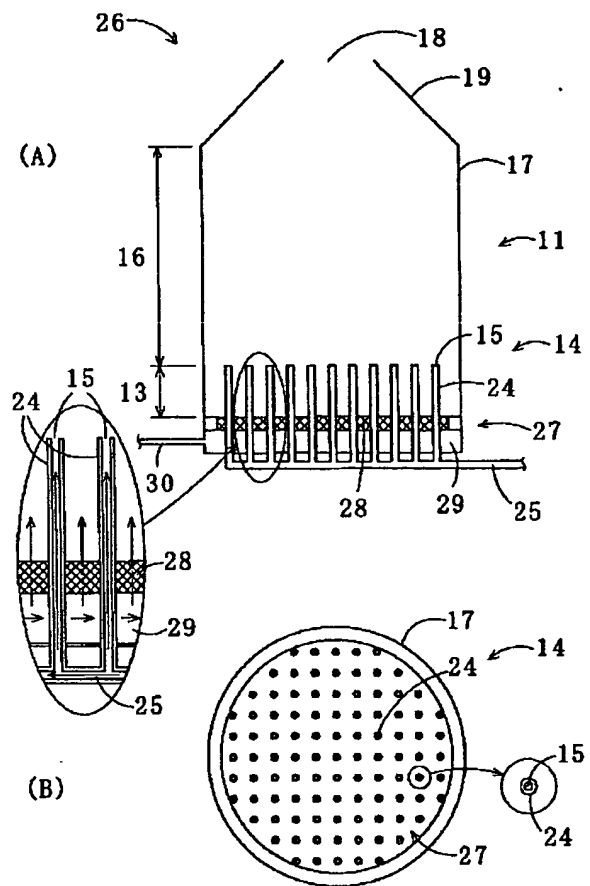
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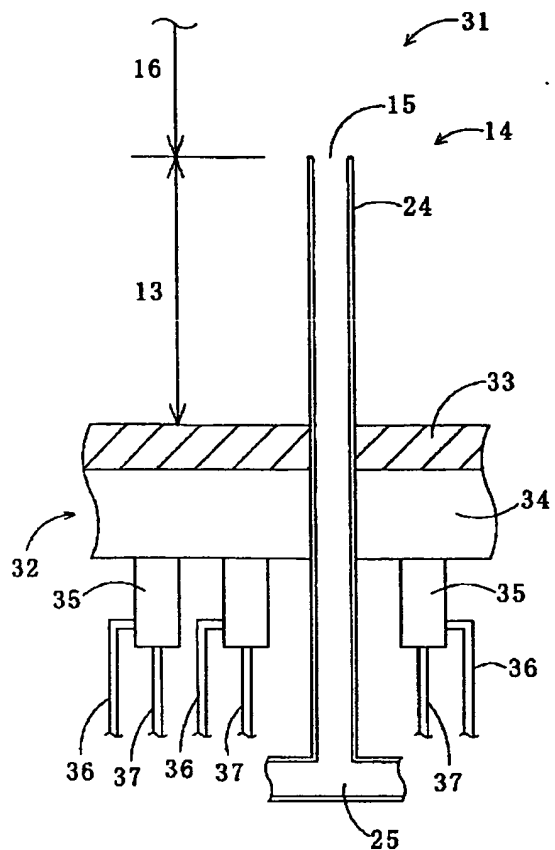
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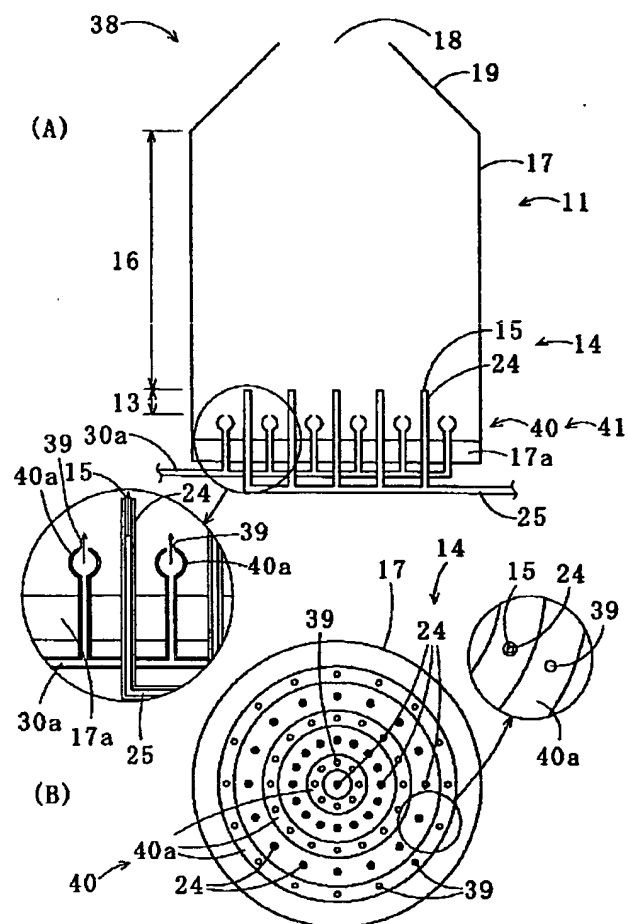
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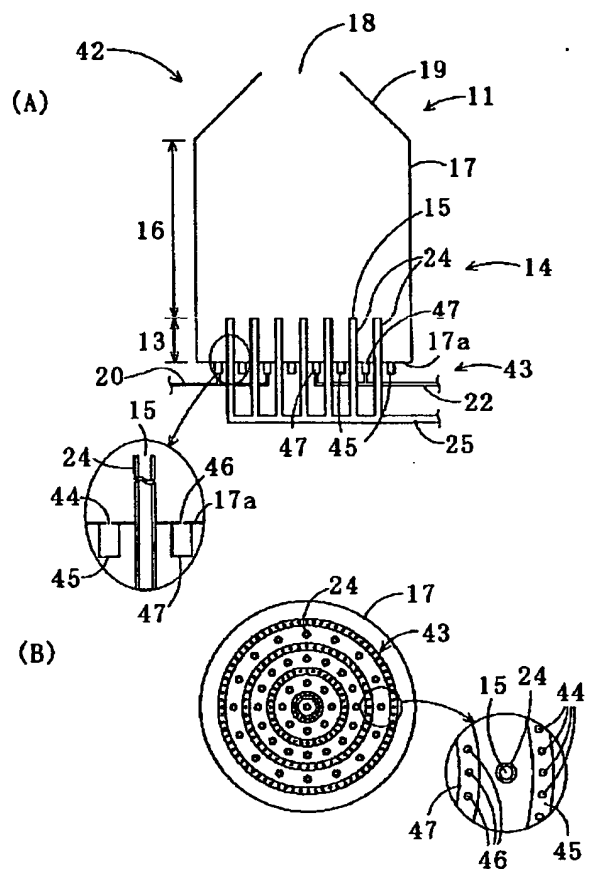
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